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CHARACTERIZATION OF SELECTED LOW—LEVEL RADIOACTIVE WASTE GENERATED BY FOUR COMMERCIAL LIGHT—WATER REACTORS

December 1977

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CHARACTERIZATION OF SELECTED LOW-LEVEL RADIOACTIVE WASTE GENERATED BY FOUR COMMERCIAL LIGHT-WATER REACTORS

BY

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White Plains, New York

through

The New York State Energy Research
and Development Authority

DECEMBER 1977

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PREFACE

The Office of Radiation Programs of the U.S. Environmental Protection Agency carries out a national program designed to evaluate population exposure to ionizing and non-ionizing radiation, and to promote development of controls necessary to protect the public health and safety. This report was prepared in order to determine the radioactivity source terms associated with the low-level wastes generated by light-water reactors and subsequently shipped to commercial shallow-land burial facilities. Readers of this report are encouraged to inform the Office of Radiation Programs of any omissions or errors. Comments or requests for further information are also invited.

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ABSTRACT

An investigation was made of the radionuclide makeup of light-water nuclear reactors' radioactive wastes presently being consigned to shallow land burial. The studies were contracted through the New York State Energy Research and Development Authority and consisted of radiochemical analyses of spent ion exchange resins, evaporator concentrates and filter sludges for specific radionuclides including activation products, fission products and transuranics. Ten waste samples were obtained from two BWRs and two PWRs.

TABLE OF CONTENTS

EPA Review Notice II
Preface.....III
Abstract.....IV
Table of Contents..... V
List of Tables.....VII
List of Figures.....VIII

TABLE OF CONTENTS

SECTION		PAGE
1.0	Introduction.	1-1
2.0	Summary	2-1
3.0	PWR and BWR Radioactive Waste Treatment Systems and Components.	3-1
3.1	General Systems Comparison.	3-1
3.2	Ion Exchange Resin Characteristics.	3-6
3.3	Evaporator Characteristics.	3-8
4.0	Waste Treatment Systems at Sampled Reactors	4-1
5.0	Radionuclide Analyses of Waste Samples Collected Under This Program.	5-1
5.1	Sample Definition and Collection Procedures	5-1
5.2	Spent Ion Exchange Resins	5-4
5.3	Evaporator Concentrates	5-7
5.4	Filter Sludges.	5-10
6.0	Radionuclide Analyses of LWR Wastes Performed Under Other Programs.	6-1
6.1	Spent Ion Exchange Resins	6-1
6.2	Evaporator Concentrates	6-2
6.3	Filter Sludges.	6-6
7.0	Comparisons, Interpretations, and Recommendations . . .	7-1
7.1	Variables Influencing Composition of Waste Sample . .	7-1
7.2	Comparison of Radionuclide Analyses	7-2
7.3	Interpretations of Data	7-7
7.4	Recommendations	7-9
8.0	Bibliography.	8-1
Appendix A -	Waste Treatment Systems at Reactors From Which Samples Were Collected.	A-1
Appendix B -	Analytical Methods Used by the Radiological Science Laboratory (RSL)	B-1

LIST OF TABLES

TABLE		PAGE
3-1	Liquid Radwaste Classification	3-2
5-1	Description of Collected Samples	5-2
5-2	Radionuclide Analysis of Spent Ion Exchange Resin Samples Measured Under This Program.	5-5
5-3	Radionuclide Analysis of Evaporator Concentrate Samples Measured Under This Program.	5-8
5-4	Radionuclide Analysis of Filter Sludge Samples Measured Under This Program.	5-11
6-1	Radionuclide Analysis of PWR Spent Resin Samples Measured Under Other Analytic Programs	6-3
6-2	Radionuclide Analysis of PWR Evaporator Concentrate Samples Measured Under Other Analytic Programs.	6-4
6-3	Radionuclide Analysis of Evaporator Concentrate Samples From Indian Point No. 2 Reactor - July, September, October, December, 1975	6-5
6-4	Radionuclide Analysis of PWR Filter Sludge Samples Measured Under Other Analytic Programs	6-7
7-1	Comparison of Concentrations of Gamma Emitting Radionuclides ($T_{\frac{1}{2}}$>300 days) In Samples of Evaporator Concentrate From PWRs	7-4
7-2	Comparison of Concentrations of Gamma Emitting Radionuclides ($T_{\frac{1}{2}}$>300 days) In Samples of Evaporator Concentrate From PWRs & BWRs.	7-5
7-3	Comparison of Concentrations of Gamma Emitting Radionuclides ($T_{\frac{1}{2}}$>300 days) In Samples of Spent Ion Exchange Resins From PWRs & BWRs	7-6

LIST OF FIGURES

FIGURE		PAGE
1	Typical System for Treatment of Liquid and Solid Radioactive Wastes at a Boiling Water Reactor.	3-4
2	Typical System for Treatment of Liquid and Solid Radioactive Wastes at a Pressurized Water Reactor.	3-5
3	Schematic Diagram of Mixed-Bed and Separate-Bed Ion Exchange Systems	3-7
4	Typical Evaporators Used in Processing Liquid Radwaste	3-9
A-1	Upgraded Liquid Radwaste System, Nine Mile Point Nuclear Station, Unit 1.	A-2
A-2	Flow Chart of the Liquid Waste System.	A-6
A-3	Liquid Waste System at R.E. Ginna.	A-10

1.0 Introduction

The purpose of this study was to provide data on the radionuclide composition and concentration in spent ion exchange resins, evaporator concentrates, and filter sludges which result from waste management operations at commercial nuclear power plants and contribute to the radioactive source term in the burial site. The characteristics of the radioactive source term are of major importance in evaluating potential movement in groundwater after emplacement in a shallow land burial site. The development of this data was to be accomplished by the analysis of systems of two PWRs and two BWRs operating in New York State, the evaluation of relevant information in the literature, and the compilation and interpretation of the available data.

The samples of BWR radwaste were obtained from the James A. Fitzpatrick and Nine Mile Point Power Stations. The PWR samples were obtained from the Indian Point 2 and R.E. Ginna Stations.

Dames & Moore reviewed the waste processing systems at the four facilities, recommended a sampling program, and compiled and analyzed the results of the radionuclide analyses performed under this study and reported in the literature.

The Radiological Science Laboratory (RSL) of the New York State Department of Health collected the samples at the reactor facilities, and performed the laboratory analyses.

2.0 Summary

This study considered the spent ion exchange resins, evaporator concentrates, and filter sludges produced at commercial nuclear power plants and disposed of by burial at shallow land burial sites. The dry solid rad-waste, which was not included, is a major contributor to the total volume of low-level waste generated, but a relatively minor contributor to the total activity in the waste. The liquid radioactive waste collection and treatment systems, in which the ion exchange units, evaporators, and filters are components, differ for BWRs and PWRs. Generalized systems for each type of reactor,and the types of liquid wastes treated,are described in Secion 3.1. In addition, the waste removal characteristics of the components are described in Section 3.2 and 3.3.

The waste treatment systems in use at the four commercial nuclear power plants at the time of sample collection were reviewed and documented. The systems are described in Section 4.0 and Appendix A. In several instances on-site modifications to the systems had been made since initial installation to improve operations.

Samples were collected at the reactors early in 1976. There were significant variations in plant operating history and the size of the samples obtained at each of the reactors. These variations are described in Section 5.1. The results of the radiometric analyses of the samples are tabulated and discussed in Sections 5.2 through 5.4. The analytic methods used to analyze the samples are described in Appendix B.

The available literature describing reactor generated waste was reviewed* so that the relevant data on radionuclide analyses of similar types of waste could be extracted and compared with the data developed under this program. This data proved to be extremely limited, and the data that was available did not include analyses for all the constituents in the waste. The radiometric data that was extracted from the literature and obtained from the plant operator's records is presented in Sections 6.2 through 6.3.

An attempt to draw definitive conclusions from the radionuclide analyses performed under this program and from prior laboratory analyses was not feasible due to the lack of a sufficient number of similar samples, and of information on the operating experience pertinent to the samples collected. However, certain analyses of selected segments of the data were made (see Section 7.2) in an attempt to determine preliminary trends. The interpretations as to radionuclide composition of the evaluated types of waste that can be supported by the available data are presented in Section 7.3.

This study does provide preliminary indications of trends of the radionuclide composition, and relative concentrations of radionuclides to be found in three types of waste generated by LWRs and disposed of at shallow land burial sites. In

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* A number of relevant references have become available since the completion of this study and, with increasing interest in this and related subjects, continue to be published. These include:

1. M.J. Steindler and L.E. Trevorrow "Wastes from the Light Water Fuel Cycle" presented at Waste Management - '76 (continued)

addition, examination of the characteristics and operating modes of the reactor waste processing system in conjunction with the analytical results provides an insight into the factors that need to be considered in developing an expanded program of sampling and analysis.

Any further confirmatory programs should be designed to permit collection of a sufficient number of samples having the same parameters so as to be statistically reliable. The parameters that need be considered are reactor and processing system characteristics, type of waste, duration of reactor operation, age of sample since generation, the location at which the sample is collected within the waste system, age of reactor and previous history. Uniformity in sample size and procedures employed in analyzing the sample need be maintained.

1.(continued) Tucson, Arizona Oct. 1976, to be published.
2. T.B. Mullarkey et. al., "A Survey and Evaluation of Handling and Disposal of Solid Low-Level Nuclear Fuel Cycle Wastes" Atomic Industrial Form, Inc. - Executive Summary Oct. 1976.

3.0 PWR and BWR Radioactive Waste Treatment Systems and Components

3.1 General Systems Comparison

In both a Pressurized Water Reactor (PWR) and a Boiling Water Reactor (BWR), the primary coolant is circulated through the reactor core and takes up heat. This, in turn, produces steam for turning the turbines which generate electrical power. The primary coolant in a BWR is the source of steam while in a PWR the primary coolant is passed through a heat exchanger and steam is produced in a secondary system. The primary coolant, in most PWRs, contains boric acid which is used as a chemical shim to control reactivity.

The primary coolant, in both a PWR and a BWR, picks up radioactive corrosion products. Additional contamination of the coolant system results from the release of fission products from defective fuel elements and diffusion of certain relatively mobile fission products (i.e. tritium) through the intact fuel element cladding. These particulates and dissolved solid contaminants are removed from the coolant stream by ion exchange resins, filters and evaporators. Other contaminated solutions generated at the reactor facility, particularly decontamination solutions, floor and laboratory drain liquids and laundry water are treated in a similar manner.

Evaporators are utilized in both BWRs and PWRs to remove those particulates and dissolved solids that are not removed or compatible to removal by ion exchange or filtration. In addition, most PWRs use evaporators to recover a portion of the boric acid. The evaporate is either reused or discharged while the concentrate is sent to the radwaste building for immobilization by incorporation into a matrix prior to packaging and

shipment for disposal.

Generalized schematics of "typical" liquid and solid radwaste treatment systems at a BWR and PWR are shown in Figure 1 and 2 respectively.

The liquid radwaste produced in nuclear power plants are categorized according to their physical and chemical properties. These categories vary between reactor types (PWR and BWR) and are shown in Table 3-1.(1) Within the reactor types, differences in design and operational features also exist.

TABLE 3-1

LIQUID RADWASTE CLASSIFICATION

PWR	BWR
Clean Wastes; low solid content liquids from controlled releases and leaks from the primary coolant loop.	High Purity Waste; liquids of low-electrical conductivity and low solids content. Primarily reactor coolant water.
Dirty Wastes; high solids content and high electrical conductivity liquids including those liquids collected from the containment buildings, auxiliary buildings and chemical laboratory.	Low Purity Wastes; Liquids of intermediate electrical conductivity. Primarily water collected from floor drains.
	Chemical Wastes; solutions of caustic and sulfuric acid which are utilized to

Blow Down Wastes; continuous or intermittent stream that is removed from the "bottoms" in the stream side of the stream generator.

Detergent Waste; includes liquids from laundry, personnel and equipment decontamination facilities.

Turbine Building Drain Waste; leakage from secondary system that is collected in the turbine building floor sump.

regenerate spent resins as well as solutions from laboratory drains and equipment drains.

Detergent Waste; laundry and personnel and equipment decontamination solutions.

A study comparing the volume and the activity of solid radwaste produced per thermal megawatt-hour of operation of BWRs and PWRs for the time period 1959-1972[2] has shown that BWRs generated a significantly higher volume of solid radwaste than PWRs, 1.50×10^{-3} ft^3 per $MW\text{-}hr_{(t)}$ and 0.56×10^{-3} ft^3 per $MW\text{-}hr_{(t)}$ respectively. However, the rate at which activity was produced was essentially the same, 3.0×10^{-5}Ci per $MW\text{-}hr_{(t)}$ for BWRs and 3.04×10^{-5}Ci per $MW\text{-}hr_{(t)}$ for PWRs. The specific activity of the PWR waste therefore is much higher than BWR wastes, 5.5×10^{-2}Ci per ft^3 to 2.03×10^{-2}Ci per ft^3 respectively.

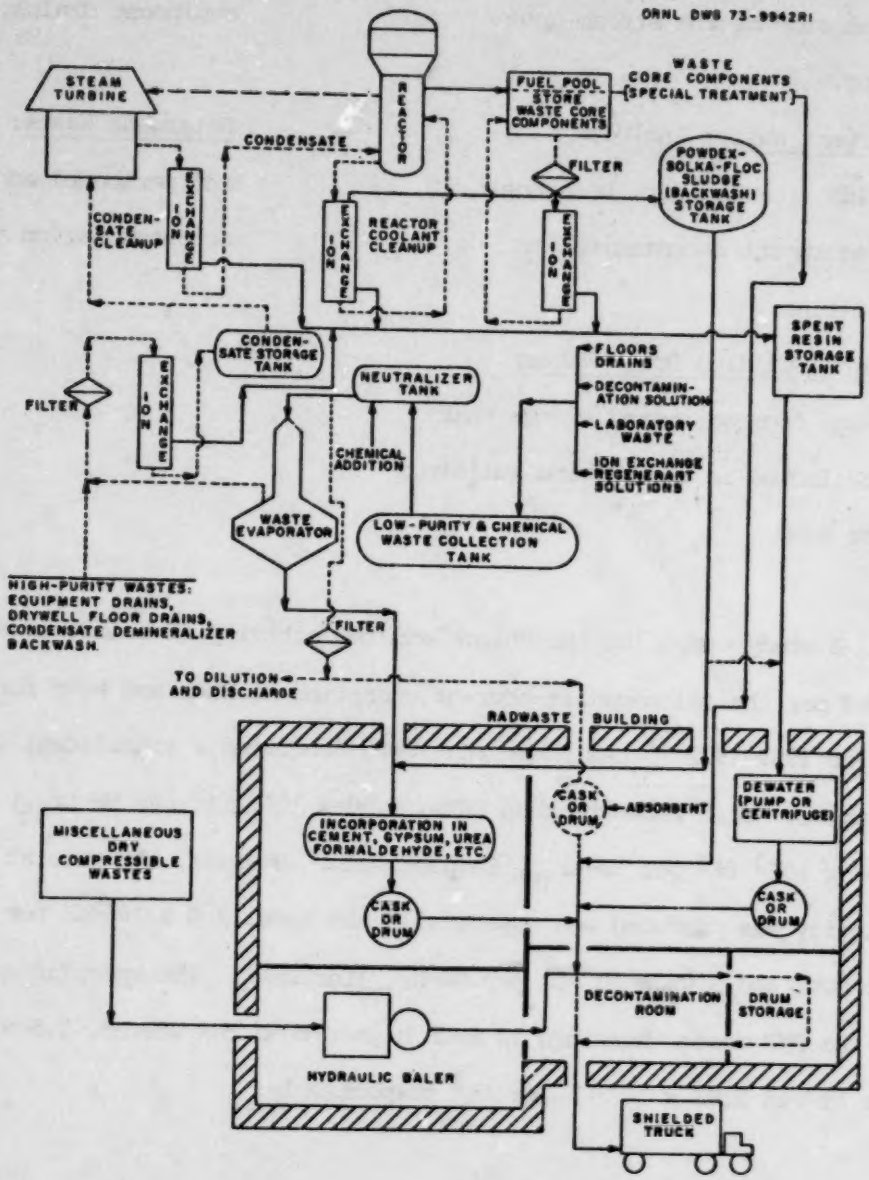


FIGURE 1

Typical System for Treatment of Liquid and Solid Radioactive Wastes at a Boiling Water Reactor (Ref.2)

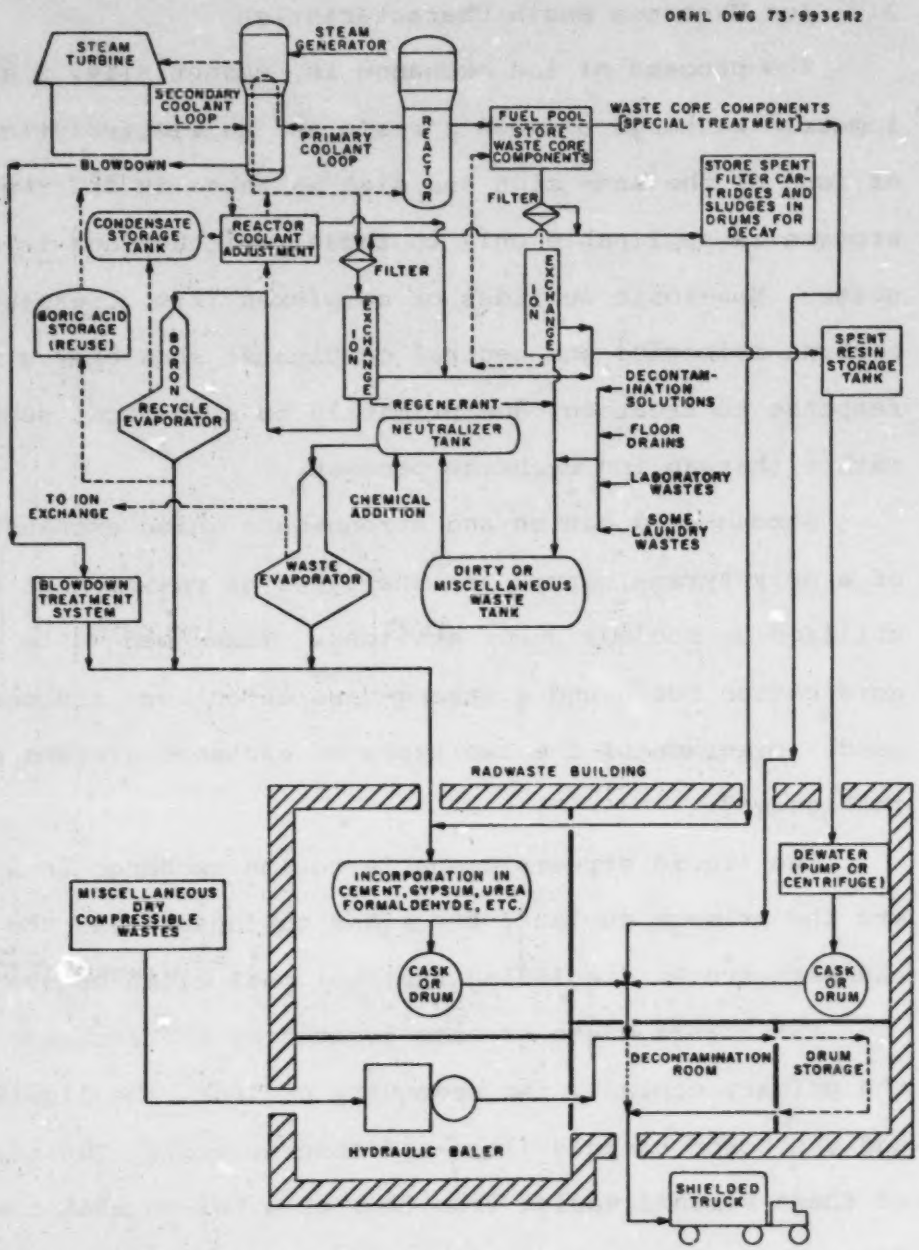


FIGURE 2

Typical System for Treatment of Liquid and Solid Radioactive Wastes at a Pressurized Water Reactor (Ref.2)

3.2 Ion Exchange Resin Characteristics

The process of ion exchange is, essentially, a stoichiometric exchange between a resin and an electrolytic solution of ions of the same sign and size as those in the resins. The process is applicable only to those radionuclides in an ionic state. Non-ionic nuclides or complexes (i.e., insoluble, neutral molecules and neutral complexes) show only a minor response to treatment due primarily to a physical sorption rather than an ion exchange process.

Strong-acid cation and strong-base anion exchange resins of a polystyrene matrix are the types of resins most frequently utilized by nuclear power stations. Mixed bed units (a strong-acid cation resin and a strong-base anion) are the most widely used. Diagrams of the two types of exchange systems are shown in Figure 3.

The liquid streams amenable to ion exchange in a BWR are the primary coolant, the steam condensate and the liquid radwaste system (including the fuel pool clean-up system).

PWR liquid waste streams treated by ion exchange include the primary coolant, the secondary coolant, the liquid radwaste and the boron recycle (feed and concentrate). The treatment of these streams varies from that of a BWR in that the letdown from the primary coolant loop is treated by both separate and mixed bed units. The boron recycle system uses a cation exchange resin.

(Ref. 3)
ORNL DWG. 72-13546

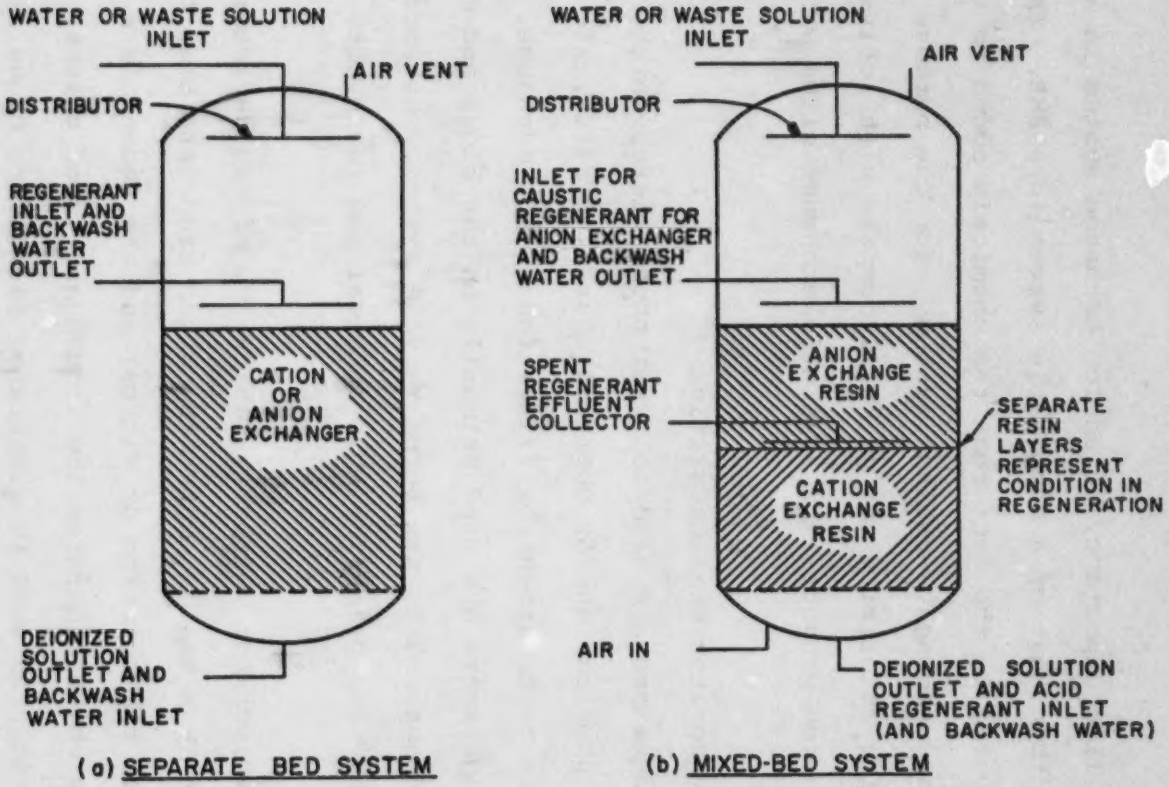

WATER OR WASTE SOLUTION INLET
AIR VENT
DISTRIBUTOR
REGENERANT INLET AND BACKWASH WATER OUTLET
CATION OR ANION EXCHANGER
DEIONIZED SOLUTION OUTLET AND BACKWASH WATER INLET
(a) SEPARATE BED SYSTEM
WATER OR WASTE SOLUTION INLET
AIR VENT
DISTRIBUTOR
INLET FOR CAUSTIC REGENERANT FOR ANION EXCHANGER AND BACKWASH WATER OUTLET
SPENT REGENERANT EFFLUENT COLLECTOR
ANION EXCHANGE RESIN
CATION EXCHANGE RESIN
SEPARATE RESIN LAYERS REPRESENT CONDITION IN REGENERATION
AIR IN
DEIONIZED SOLUTION OUTLET AND ACID REGENERANT INLET (AND BACKWASH WATER)
(b) MIXED-BED SYSTEM

Fig. 3. Schematic Diagram of Mixed-Bed and Separate-Bed Ion Exchange Systems.

The radionuclides removed from the PWR waste streams by ion exchange are essentially the same as those removed in a BWR.

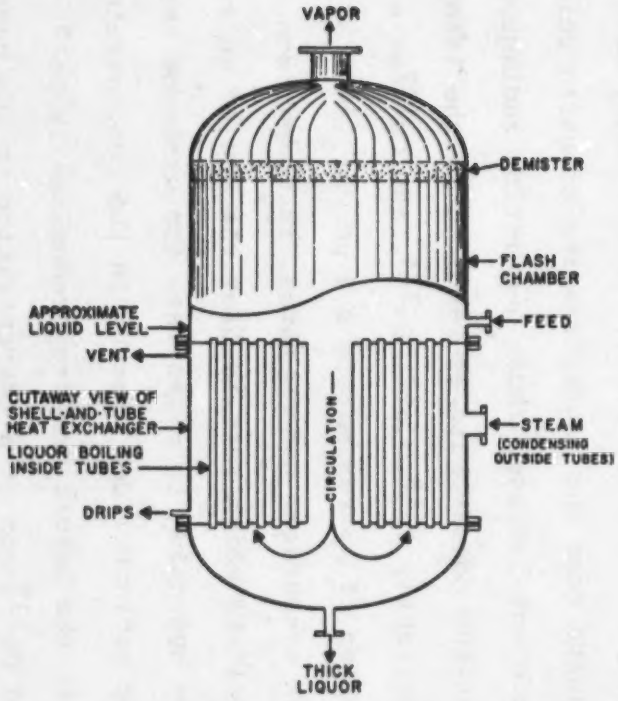
The life expectancy of an ion exchange system in a PWR is lower than that of a comparable system in a BWR. This is attributed to the fact that the chemicals added to the primary and secondary coolant systems, for the purpose of controlling reactivity and pH, will compete with activation and corrosion products for available exchange sites within the resin.

3.3 Evaporator Characteristics

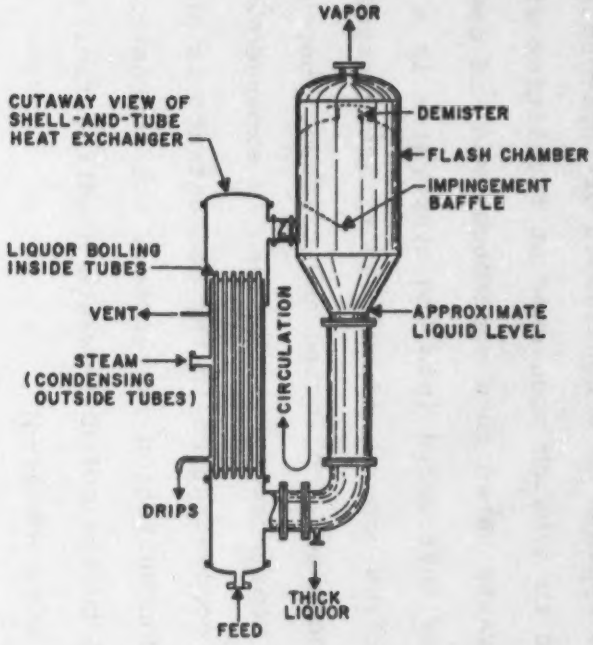
Evaporators are used to treat those wastes which, due to their physical and/or chemical characteristics, are not compatible to treatment by filtration or ion exchange. In PWRs, evaporators are used primarily on the clean and dirty waste streams and in the boron recycle system. Evaporators in BWRs handle, primarily, the chemical and low purity waste streams.

An evaporator consists, basically, of two devices; the first is a heating apparatus which transfers heat for boiling to the solution or slurry; and the second is a mechanism which separates the liquid and vapor phases. The basic principles used in evaporator design are those of heat transfer, vapor-liquid separation, volume reduction and energy utilization.[1] Diagrams of the two most commonly utilized types of radwaste evaporators are shown in Figure 4.

(Ref 1)
ORNL DWG. 73-8589 R1



CALANDRIA-TYPE EVAPORATOR



LONG-TUBE RECIRCULATION EVAPORATOR

Fig. 4 Typical Evaporators Used in Processing Liquid Radwaste.

Under routine operating conditions, these radwaste evaporators operate on a continuous or semi-continuous mode as compared to a batch mode used at facilities with a low volume of waste. When in a continuous mode of operations, the waste is introduced into the evaporator in a predetermined volume, boiling occurs and the vapors are continuously removed, condensed, collected and treated. The evaporation process is continued until the feed is expended or a predetermined concentration in the concentrate is obtained. Once this concentration is reached, the concentrate is transferred to the solid radwaste handling facility for processing and packaging.

PWR evaporator concentrates, excluding the concentrate from the boron recovery system, are primarily sodium borate which results from the neutralization of boric acid from primary coolant leakage. Those γ-emitting radionuclides present in the concentrate as reported in the literature are predominantly Co^{58}, Co^{60}, Cs^{134}, and Cs^{137}at a total concentration of approximately 0.2 μCi/ml.(4)

BWR evaporator concentrates, in comparison, are primarily soldium sulfate which results from the use of sulfuric acid and sodium hydroxide to regenerate ion exchange resins. The γ-emitting radionuclides present in the concentrate as reported in the literature are predominantly Co^{58}, Co^{60}, Cs^{134}, and Cs^{137} and at a concentration in the range of 2.0-3.0 μCi/ml.(4)

4.0 Waste Treatment Systems at Sampled Reactors

The liquid radwaste systems at a commercial nuclear power plant are designed to collect, monitor and process for reuse or disposal, all potentially radioactive liquid wastes. The residues of the processing of the liquid waste streams, are evaporator concentrates, filter sludges, and spent ion exchange resins. These materials are immobilized by different techniques, packages and shipped to a burial site for disposal.

The nuclear power plants participating in this study provided representative radwaste samples from both PWR and BWR systems having a range of operating lifetimes. The power plants sampled were;

(1) Indian Point No. 2- A PWR operated by the Consolidated Edison Company having a net capacity of 873 MWe that began commercial operation in August, 1973.

(2) R.E. Ginna- A PWR operated by the Rochester Gas & Electric Company having a net capacity of 420 MWe that began commercial operation in July, 1970.

(3) Nine Mile Point- A BWR operated by the Niagara Mohawk Power Corporation having a net capacity of 610 MWe that began commercial operation in December, 1969.

(4) James A. Fitzpatrick- A BWR operated by the Power Authority of the State of New York having a capacity of 821 MWe that began commercial operation in July, 1975.

The liquid and solid radwaste systems used at these four power plants at the time of sample collection were reviewed from the available

literature (5,7,8,9,11) and through personal visits to each of the plants and conversations with the knowledgeable plant personnel. (6,10,12) In several instances, the systems had been modified from the published descriptions. The radwaste systems at each of the facilities are described in Appendix A.

5.0 Radionuclide Analyses of Waste Samples Collected During This Program

5.1 Sample Definition and Collection Procedures

The program provided funds for the collection and analysis of ten (10) samples distributed among four (4) reactors. This approach, it was considered, would provide analyses of the waste constituents from two (2) PWRs and two (2) BWRs of varying periods of accumulated operating time since start up, and permit comparison of the differences in radionuclide concentrations resulting from these factors.

The reactor facilities were visited by Dames & Moore personnel and discussion held with plant personnel to determine the accessibility of the waste processing and packaging, and the availability of each type of sample. Based on the information obtained, a sampling program was recommended consisting of two (2) evaporator concentrate samples from each facility. Resin samples and filter sludges would not be collected because the reactor operators had indicated that these samples could not readily be made available. When the Radiological Science Laboratory (RSL) collected the samples at the facilities, it was necessary to revise this program because the reactor operators were able to make available certain filters and resin samples and could not provide all the specified evaporator concentrate samples. The samples collected from each reactor, and the conditions under which they were collected, are described in Table 5-1.

TABLE 5-1

Description of Collected Samples [15]

Reactor	Waste Type	Description
R.E. Ginna	Evaporator Concentrate	<20 ml sample collected; high undissolved solids and salts content which hampered titration; small sample size and solids content prevented volumetric conversion and required reporting of results on a weight basis.
	Filter Sludge	Sample consisted of 3 surface smears of the Primary Coolant Filters which had been in-line for approximately 1 year; Station Health Physicist considered collection of an actual filter sample to be inadvisable due to >100mr potential personnel exposures; nature of sample required reporting of results on a per filter basis.
	Spent Resin	~1 ml sample of resin beads (wet) collected from spent resin storage tank; length of time resin in-line is unknown; results reported on a weight basis because volume of beads could not readily be measured.
Indian Point No. 2	Evaporator Concentrate	25 ml sample collected from composite evaporator in Station No.1 and diluted to 500 ml; results expressed on volumetric basis.
	Filter Sludge	Sample obtained from filter placed in the tap line of the primary coolant system thru which 304 liters of primary coolant was passed; Station Health Physicist considered collection of an actual filter sample to be inadvisable due to >100mr potential exposures; results reported on a per filter basis.
	Spent Resin	No sample collected.

TABLE 5-1 (cont'd.)

Reactor	Waste Type	Description
Nine Mile Point	Evaporator Concentrate	~1 ml sample of unknown age and which had been previously collected and stored at site; high, undissolved solids and salt content which hampered titration; results reported on a weight basis.
	Filter Sludge	~1 ml sample of unknown age collected from the sludge storage tank; results reported on a weight basis.
	Spent Resin	~1 ml sample of resin beads (dry) collected from spent resin storage tank; length of time resin in-line is unknown; results reported on a weight basis.
James A. Fitzpatrick	Evaporator Concentrate	1 liter sample collected; solids content unknown; results reported on volumetric basis.
	Filter Sludge	~45 ml of dry centrifuge waste (powder) collected; results reported on weight basis.
	Spent Resin	No sample collected.

As can be noted from examination of Table 5-1, the samples of each type of waste collected varied in size, prior history, and in the case of the filter sludge, in the type of sample collected. Thus many of the factors that may influence the radionuclide composition of the waste types vary from sample to sample, making a comparison among sample analyses difficult. The comparative analyses that can be made between similar waste types from the different reactors are provided in Section 7.0.

The analytical procedure employed by RSL to analyze the samples of each type of waste collected are described in Appendix B for the various radionuclides evaluated.

5.2 Spent Ion Exchange Resins

The results of the radiometric analyses of spent ion exchange resins performed by RSL are presented in Table 5-2. Both the concentrations and the relative percent of the individual radionuclides in each sample are provided. Samples were available from only the Nine Mile Point (BWR) and R.E. Ginna (PWR) facilities.

Although the percentages of the radionuclides present vary between the two samples, in each instance three of the radionuclides, Cs^{137}, Cs^{134} and Co^{60}, account for approximately 90% of the total concentration, with Cs^{137} being the predominant radionuclide in both samples. The concentrations of each sample are quite comparable, 43.23μ Ci/gm for Nine Mile Point, and 41.03μ Ci/gm for Ginna.

TABLE 5-2

Radionuclide Analysis of Spent Ion Exchange Resin Samples Measured Under This Program

Radionuclide		Nine Mile Point		R.E. Ginna	
		Concentration (μCi/gm)	Relative Proportion (%)	Concentration (μCi/gm)	Relative Proportion (%)
Am^{241}	(f.p.)	6×10^{-5}	<.1	7×10^{-4}	<.1
$Pu^{239,240}$	(f.p.)	3×10^{-5}	<.1	8×10^{-4}	<.1
Pu^{238}	(f.p.)	4×10^{-5}	<.1	4×10^{-4}	<.1
U^{238}	(f.p.)	$<3 \times 10^{-6}$	<.1	4.5×10^{-5}	<.1
U^{235}	(f.p.)	$<5 \times 10^{-6}$	<.1	$<1.2 \times 10^{-5}$	<.1
U^{234}	(f.p.)	$<7 \times 10^{-6}$	<.1	2.3×10^{-5}	<.1
Ce^{144}	(f.p.)	0.12	0.3	0.3	0.7
Cs^{137}	(f.p.)	31.7	73.4	21.9	53.4
Cs^{134}	(f.p.)	2.9	6.7	12.4	30.2
I^{131}	(f.p.)	ND	----	ND	----
I^{129}	(f.p.)	$<2 \times 10^{-6}$	<.1	6×10^{-4}	<.1
Sb^{125}	(f.p.)	0.11	0.3	0.2	0.5
Sb^{124}	(a.p.)	ND	----	ND	----
Ru^{106}	(f.p.)	0.3	0.7	0.7	1.7
Tc^{99}	(f.p.)	9×10^{-3}	<.1	2×10^{-3}	<.1
Zr^{95}	(f.p. or a.p.)	ND	----	ND	----
Nb^{95}	(f.p. or a.p.)	ND	----	ND	----
Sr^{90}	(f.p.)	7.2×10^{-2}	0.2	8.5×10^{-2}	0.2
Zn^{65}	(a.p.)	8×10^{-2}	0.2	0.15	0.4
Ni^{63}	(a.p.)	9.7×10^{-3}	<.1	1.39	3.4
Co^{60}	(a.p.)	6.24	14.5	2.06	5.0
Fe^{59}	(a.p.)	ND	----	ND	----

TABLE 5-2 (cont'd.)

Radionuclide		Concentration (μCi/gm)	Relative Propertion (%)	Concentration (μCi/gm)	Relative Proportion (%)
Co^{58}	(a.p.)	0.36	0.8	0.5	1.2
Co^{57}	(a.p.)	ND	----	ND	----
Fe^{55}	(a.p.)	0.174	0.4	1.01	2.5
Mn^{54}	(a.p.)	9×10^{-2}	0.2	0.16	0.4
Cr^{51}	(a.p.)	ND	----	ND	----
$C^{14}_{(CO_2)}$	(a.p.)	2×10^{-4}	<.1	5×10^{-3}	<.1
C^{14} (CH_{14})	(a.p.)	$<8 \times 10^{-6}$	<.1	5×10^{-6}	<.1
H^3	(f.p. or a.p.)	3×10^{-3}	<.1	0.125	0.3
Total Concentration		43.23		41.03	

ND - Not Detected

f.p. - fission product

a.p. - activation product

It should be noted that the major constituents delineated in this and succeeding sections on the basis of measured concentration in the samples will not necessarily be the major constituents remaining over the long term after radionuclide decay has occurred.

5.3 Evaporator Concentrates

The results of the radiometric analyses of the evaporator concentrate samples are presented in Table 5-3. The concentrations and relative percent of each radionuclide are provided. Samples were analyzed from all four of the reactors.

The radionuclide concentration vary from sample to sample within each reactor type, and between reactors. In the sample from Indian Point No. 2, the major constituents in order of predominance are I^{131}, Cs^{137}, Cs^{134}, Co^{58}, and Fe^{55}, comprising approximately 90% of the total concentration. In the evaporator sample from Ginna, H^{3}, Cs^{137}, Cs^{134}, Co^{58}, and Co^{60}, in that order, are the major constituents, comprising approximately 95% of the total concentration. In the case of Nine Mile Point, Fe^{55}, Cs^{137}, Cs^{134}, Co^{60}, Mn^{54} represent 95% of the total concentration of the sample. While in the sample from Fitzpatrick the major constituents in order of predominance are Mn^{54}, Co^{58}, Co^{60}, Cr^{51}, and Zn^{65} comprising approximately 87% of the total concentration.

Table 5-3

Radionuclide Analysis of Evaporator Concentrate Samples
Measured under this Program

Radionuclide	Indian Point No. 2		R.E Ginna		Nine Mile Point		J.A. Fitzpatrick	
	Concentration (µCi/ml)	Relative Proportion (%)	Concentration (µCi/gm)	Relative Proportion (%)	Concentration (µCi/gm)	Relative Proportion (%)	Concentration (µCi/ml)	Relative Proportion (%)
Am^{241}	3.0×10^{-7}	<.1	ND	----	5.0×10^{-6}	<.1	ND	----
$Pu^{239,240}$	8.0×10^{-8}	<.1	1.8×10^{-6}	<.1	8.0×10^{-6}	<.1	5.5×10^{-8}	<.1
Pu^{238}	2.0×10^{-7}	<.1	1.0×10^{-6}	<.1	1.3×10^{-5}	<.1	1.6×10^{-8}	<.1
U^{238}	1.9×10^{-7}	<.1	1.88×10^{-6}	<.1	1.5×10^{-6}	<.1	7.0×10^{-9}	<.1
U^{235}	8.0×10^{-8}	<.1	2.0×10^{-7}	<.1	2.0×10^{-6}	<.1	1.0×10^{-8}	<.1
U^{234}	1.2×10^{-7}	<.1	3.0×10^{-7}	<.1	3.0×10^{-6}	<.1	1.6×10^{-8}	<.1
Ce^{144}	ND	----	8.0×10^{-4}	0.2	6.0×10^{-3}	0.7	2.0×10^{-4}	0.4
Cs^{137}	0.3	21.8	0.102	29.9	0.229	27.0	4.0×10^{-4}	0.9
Cs^{134}	0.19	13.8	3.7×10^{-2}	10.8	0.169	19.9	1.0×10^{-4}	0.2
I^{131}	0.41	29.8	ND	----	ND	----	ND	----
I^{129}	2.0×10^{-5}	<.1	4.0×10^{-6}	<.1	1.0×10^{-4}	<.1	4.0×10^{-7}	<.1
Sb^{125}	ND	----	1.0×10^{-2}	0.3	6.0×10^{-3}	0.7	2.0×10^{-4}	0.2
Sb^{124}	ND	----	1.0×10^{-4}	<.1	ND	----	4.0×10^{-4}	0.9
Ru^{106}	0.007	0.5	2.0×10^{-3}	0.6	1.9×10^{-2}	2.2	8.0×10^{-4}	1.8
Tc^{99}	2.0×10^{-5}	<.1	7.0×10^{-5}	<.1	1.0×10^{-3}	0.1	1.6×10^{-6}	<.1
Zr^{95}	ND	----	6.0×10^{-4}	<.1	ND	----	5.0×10^{-4}	1.1

TABLE 5-3 (cont'd)

Radionuclide	Indian Point No. 2		R.E. Ginna		Nine Mile Point		J.A. Fitzpatrick	
	Concentration (μCi/ml)	Relative Proportion (%)	Concentration (μCi/gm)	Relative Proportion (%)	Concentration (μCi/gm)	Relative Proportion (%)	Concentration (μCi/ml)	Relative Proportion (%)
Nb^{95}	ND	----	7.0×10^{-4}	.1	ND	----	9.0×10^{-4}	2.0
Sr^{90}	7.0×10^{-5}	<.1	7.6×10^{-5}	.1	1.3×10^{-3}	0.2	7.0×10^{-7}	.1
Zn^{65}	ND	----	3.0×10^{-4}	.1	4.0×10^{-3}	0.5	3.4×10^{-3}	7.6
Ni^{63}	1.91×10^{-2}	1.4	6.1×10^{-3}	1.8	2.2×10^{-3}	0.3	1×10^{-4}	0.2
Co^{60}	3.5×10^{-2}	2.5	1.89×10^{-2}	5.5	9.6×10^{-2}	11.3	8.9×10^{-3}	19.9
Fe^{59}	3.0×10^{-3}	0.2	ND	----	ND	----	ND	----
Co^{58}	0.1890	13.7	3.46×10^{-2}	10.1	ND	----	$.09 \times 10^{-2}$	24.4
Co^{57}	3.0×10^{-4}	<.1	1.2×10^{-4}	<.1	5×10^{-4}	.1	3×10^{-5}	<.1
Fe^{55}	0.1280	9.3	4.4×10^{-3}	1.3	0.2900	34.1	7×10^{-4}	.1
Mn^{54}	3.1×10^{-2}	2.3	1.0×10^{-3}	0.3	2.3×10^{-2}	2.7	1.18×10^{-2}	26.4
Cr^{51}	3.64×10^{-2}	2.6	ND	----	ND	----	3.7×10^{-3}	8.3
$C^{14}_{(CO_2)}$	2.1×10^{-5}	<.1	6×10^{-5}	<.1	1.8×10^{-6}	<.1	7.1×10^{-6}	<.1
$C^{14}_{(CH_4)}$	2.1×10^{-7}	<.1	4.0×10^{-7}	----	9.0×10^{-7}	<.1	6×10^{-9}	<.1
H^3	2.72×10^{-2}	1.97	0.132	38.6	2.5×10^{-3}	0.3	1.7×10^{-3}	3.8
Total Concentration	1.3759		0.3417		0.8492		0.0447	

5.4 Filter Sludges

The results of the radiometric analyses of the filter sludge or selected "equivalent" samples are presented in Table 5-4. The data is here again reported as concentrations and relative percent of each radionuclide of the total concentration for each sample. Samples, of varying origin (see Table 5-1), were collected from each of the reactors.

Again substantial variation in relative concentrations of the various radionuclides can be noted among the four samples. In the filter sludge sample from Indian Point No. 2 the major constituents in order of predominance are Cr^{51}, Co^{58}, and Co^{60} which together comprise in excess of 95% of the total specific activity of the sample. In the analysis of the surface smears of the primary coolant filter from Ginna, containing sludge particles representative of the material collected for packaging, Fe^{55}, Co^{60}, and Ni^{63} in that order are the major constituents comprising approximately 86% of the total specific activity. In the sample from the sludge storage tank at Nine Mile Point the major constituents are Fe^{55}, Cs^{137}, and Cs^{134} comprising approximately 88% of the total specific activity of the sample. And in the powdered dry centrifuge waste sample from Fitzpatrick, Co^{57}, Mn^{54}, Fe^{55} and Co^{58} comprise approximately 88% of the activity in the sample. As in the case of the evaporator samples, the various types of filter sludge samples exhibit a wide range in total activity.

Table 5-4

Radionuclide Analysis of Filter Sludge
Samples Measured Under This Program

Radionuclide	Indian Point No. 2		R.E. Ginna		Nine Mile Point		J.A. Fitzpatrick	
	Concentration (μCi/filter)	Relative Proportion (%)	Concentration (μCi/filter)	Relative Proportion (%)	Concentration (μCi/gm)	Relative Proportion (%)	Concentration (μCi/gm)	Relative Proportion (%)
Am^{241}	ND	----	3.07×10^{-4}	<.1	1.8×10^{-5}	<.1	2.5×10^{-6}	<.1
$Pu^{239,240}$	5.5×10^{-5}	<.1	5.9×10^{-4}	<.1	1.5×10^{-4}	<.1	5.0×10^{-7}	<.1
Pu^{238}	1.3×10^{-5}	<.1	2.35×10^{-4}	<.1	2.8×10^{-4}	<.1	1.2×10^{-6}	<.1
U^{238}	3.0×10^{-6}	<.1	6.0×10^{-7}	<.1	2.0×10^{-5}	<.1	1.9×10^{-6}	<.1
U^{235}	4.0×10^{-6}	<.1	4.0×10^{-7}	<.1	1.8×10^{-5}	<.1	5.0×10^{-7}	<.1
U^{234}	6.0×10^{-6}	<.1	2.6×10^{-6}	<.1	3.0×10^{-5}	<.1	7.0×10^{-7}	<.1
Ce^{144}	3.0×10^{-2}	0.1	1.4×10^{-2}	0.9	6×10^{-2}	0.5	2×10^{-3}	.1
Cs^{137}	0.1510	0.6	4.1×10^{-3}	0.3	1.130	9.0	8×10^{-4}	<.1
Cs^{134}	0.1260	0.5	9×10^{-4}	<.1	0.7400	5.9	8×10^{-4}	<.1
I^{131}	ND	----	ND	----	ND	----	ND	----
I^{129}	8.0×10^{-5}	<.1	1.8×10^{-6}	<.1	1.1×10^{-4}	<.1	3.0×10^{-6}	<.1
Sb^{125}	3.9×10^{-2}	0.2	4.1×10^{-3}	0.3	4×10^{-2}	0.3	1.9×10^{3}	0.1
Sb^{124}	ND	----	ND	----	ND	----	ND	----
Ru^{106}	0.1	0.4	3.9×10^{-2}	2.5	8×10^{-2}	0.6	1.1×10^{-3}	<.1
Tc^{99}	1.4×10^{-3}	<1	9×10^{-4}	<1	8×10^{-2}	<.1	6×10^{-5}	<.1
Zr^{95}	5.3×10^{-2}	0.2	1.3×10^{-2}	0.8	ND	----	6×10^{-3}	0.3

TABLE 5-4 (cont'd.)

Radionuclide	Indian Point No. 2		R.E. Ginna		Nine Mile Point		J.A. Fitzpatrick	
	Concentration (µCi/filter)	Relative Proportion (%)	Concentration (µCi/filter)	Relative Proportion (%)	Concentration (µCi/gm)	Relative Proportion (%)	Concentration (µCi/gm)	Relative Proportion (%)
Nb^{95}	0.1090	0.5	2.38×10^{-2}	1.5	ND	----	1.13×10^{-2}	0.6
Sr^{90}	7×10^{-4}	<.1	1×10^{-2}	0.6	5.7×10^{-2}	0.5	$<1 \times 10^{-4}$	<.1
Zn^{65}	8.5×10^{-2}	0.4	2.5×10^{-3}	0.2	6.9×10^{-2}	0.6	3.1×10^{-2}	1.6
Ni^{63}	1.1×10^{-2}	<.1	9.3×10^{-2}	6.0	2.8×10^{-2}	0.2	7.4×10^{-3}	0.4
Co^{60}	1.9100	8.4	0.2550	16.5	1.5300	12.2	0.1230	6.4
Fe^{59}	0.1880	0.8	ND	----	ND	----	2.9×10^{-2}	1.5
Co^{58}	9.4000	40.7	3.9×10^{-2}	2.5	6.4×10^{-2}	0.5	0.262	13.8
Co^{57}	1.9×10^{-2}	<.1	4×10^{-4}	<.1	2×10^{-3}	<.1	0.800	41.8
Fe^{55}	4.38×10^{-2}	0.2	0.9800	63.4	7.7000	61.4	0.28	14.6
Mn^{54}	0.3230	1.4	1.25×10^{-2}	0.7	0.5300	4.2	0.346	18.1
Cr^{51}	10.5000	45.9	1.2×10^{-2}	0.8	0.5000	4.0	ND	----
$C^{14}_{(CO_2)}$	2.36×10^{-2}	0.1	3.9×10^{-2}	2.5	1×10^{-3}	<.1	1×10^{-4}	<.1
$C^{14}_{(CH_4)}$	7×10^{-7}	<.1	8.0×10^{-8}	<.1	7.0×10^{-5}	<.1	2.5×10^{-7}	<.1
H^3	ND	----	1.3×10^{-3}	0.1	2.0×10^{-3}	<.1	ND	----

6.0 Radionuclide Analyses of LWR Waste Performed Under Other Programs

The limited amount of available literature containing specific analyses of similar waste types generated at LWRs was reviewed to determine the extent of available data that could be used as a basis for comparison with the data developed under this program. The review revealed that the amount of relevant data is limited, and that which is available does not provide a complete breakdown of all the constituents to be found in the various waste forms. Most of the reported analyses of waste, as concentrates, filter sludge, or resins, or as solidified wastes, were of gross activity levels measured to assure that the plant systems were continuing to operate within specifications and that no anomalies were present.

Of the available semi-annual Effluent and Waste Disposal Reports prepared for the four reactors whose wastes were analyzed under this study, only those for Ginna [13] provided any relevant radionuclide breakdown.

The data of relevance to this study that was available from prior work is presented in the succeeding sections. It was obtained both from published reports [13,14] and from the plant operators records.[12] The data is limited to waste generated at PWRs; comparable BWR data is not available in the literature.

6.1 Spent Ion Exchange Resins

The results of the radiometric analyses of spent resin samples performed by the laboratory personnel at Ginna [13] and Indian Point

No. 2 [12] and that reported for an unnamed PWR [14] are presented in Table 6-1. The analyses are limited to certain significant gamma emitting radionuclides which, it can be noted, vary considerably among the samples analyzed. Both the concentrations (or total activities) and relative percent of the individual radionuclides analyzed in each sample are provided.

6.2 Evaporator Concentrates

The results of the radiometric analyses of evaporator concentrates from the same three reactors reported on for spent resin constituents are presented in Table 6-2. The analyses are similarly limited, covering only certain gamma emitting radionuclides which also show substantial variation from sample to sample.

Data available from monthly analyses of evaporator concentrate samples collected from the Indian Point No. 2 reactor during four of the months in the period from July to December 1975 provides an indication of the variation of concentrations of certain of the radionuclides with time. The concentrations of the measured radionuclides and their relative proportion of the total activity are presented in Table 6-3.

The concentration of each constituent in the total sample vary during the six month sampling period, (corresponding to semi-annual reporting period for the reactors), but no consistent pattern can be found. For example, the concentration of Cs^{134} showed a variation greater than 30 fold during this period, while the concentration of

TABLE 6-1

Radionuclide Analysis of PWR Spent Resin Samples
Measured Under Other Analytic Programs

Radionuclide	Reactor					
	RE Ginna[13]		Indian Pt.2[12]		Unidentified PWR[14]	
	Concentration (μCi/ml)	Relative Proportion* (%)	Activity (Ci)	Relative Proportion* (%)	Concentration (μCi/ml)	Relative Proportion* (%)
Cs^{137}	28.23	30.2	5.77	72.0	14.32	47.6
Cs^{134}	7.38	7.9	1.31	16.0	3.89	12.9
Sb^{125}	N.R.	----	N.R.	----	0.137	0.5
Co^{60}	37.29	39.9	0.91	11.0	5.23	17.4
Co^{58}	N.R.	----	N.R.	----	3.51	11.7
Mn^{54}	20.47	21.9	0.03	1.0	2.97	9.9

*of only those radionuclides analyzed.

TABLE 6-2

Radionuclide Analysis of PWR Evaporator Concentrate Samples
Measured Under Other Analytic Programs

Radionuclide	Reactor					
	RE Ginna[13]		Indian Pt.2[12]		Unidentified PWR[14]	
	Concentration (µCi/ml)	Relative Proportion* (%)	Concentration** (µCi/ml)	Relative Proportion* (%)	Concentration (µCi/ml)	Relative Proportion* (%)
Cs^{137}	0.542	60.4	0.718	43.3	0.000532	7.0
Cs^{134}	0.272	30.3	0.532	28.5	N.R.	---
I^{131}	0.013	1.3	N.R.	----	N.R.	---
Co^{60}	0.045	5.0	0.020	2.6	0.00599	79.3
Co^{58}	N.R.	----	0.388	22.2	0.000523	6.9
Mn^{54}	0.021	2.2	0.014	1.4	0.000514	6.8

*of only those radionuclides analyzed.
**average concentration July-December 1975.

NR = Not Reported

TABLE 6-3

Radionuclide Analysis of Evaporator Concentrate Samples From Indian Point No. 2 Reactor
July, September, October, December, 1975

Radionuclide	Month							
	July		September		October		December	
	Concentration (μCi/ml)	Relative Proportion* (%)	Concentration (μCi/ml)	Relative Proportion* (%)	Concentration (μCi/ml)	Relative Proportion* (%)	Concentration (μCi/ml)	Relative Proportion* (%)
Cs^{137}	2.350	55.5	0.169	40.4	0.249	42.5	0.105	34.8
Cs^{134}	1.810	42.8	0.113	27.0	0.149	25.4	0.057	18.8
Co^{60}	0.017	0.4	0.017	4.1	0.024	4.0	0.020	6.6
Co^{58}	0.019	0.5	0.113	27.0	0.147	25.1	0.109	36.1
Mn^{54}	0.020	0.5	0.007	1.6	0.017	3.0	0.011	3.6

*of only those radionuclides analyzed.

Co^{60} remained essentially constant. On a relative basis, the concentrations in each sample also tended to show an inconsistent pattern of variation. For the same two radionuclides, Cs^{134} ranged from a high of 43% of the total sample measured to a low of 19%, while Co^{60} ranged from a high of almost 7% to a low of less than 1%.

6.3 Filter Sludges

The results of the radiometrc analyses of filter sludges collected from Indian Point No. 2 [12] and from the unnamed PWR [14] are presented in Table 6-4. The filter samples analyzed from the latter include samples from the spent fuel pool, the reactor coolant system, and the waste holding tank, while the sample from Indian Point No. 2 is from a single unidentified location in the system. The tabulation shows both the concentration and relative percent of the individual radionuclide in each sample.

TABLE 6-4

Radionuclide Analysis of PWR Filter Sludge Samples Measured
Under Other Analytic Programs

Radionuclide	Indian Pt.2[12]		Unidentified PWR[14]					
	RCS		SFP		RCS		WHT	
	Concentration (μCi/gm)x10^3	Relative Proportion (%)	Concentration (μCi/gm)	Relative Proportion (%)	Concentration (μCi/gm)	Relative Proportion (%)	Concentration (μCi/gm)	Relative Proportion (%)
Sb^{125}	N.R.	----	0.581	1.2	N.R.	----	.0124	1.1
Cd^{115m}	N.R.	----	7.750	16.6	N.R.	----	N.R.	----
Cd^{113m}	N.R.	----	0.049	0.1	N.R.	----	N.R.	----
Ag^{110m}	N.R.	----	0.327	0.7	1.851	4.7	.0144	1.3
Zr^{95}	N.R.	----	0.131	0.3	N.R.	----	N.R.	----
Zn^{65}	N.R.	----	N.R.	----	N.R.	----	.0099	0.9
Co^{60}	1.89	15.1	2.490	5.3	6.80	17.1	.509	47.0
Co^{58}	3.65	29.2	19.4	41.8	28.6	72.2	.464	42.9
Co^{57}	N.R.	----	0.036	0.1	0.109	0.3	.0021	0.2
Mn^{54}	0.362	2.9	1.32	2.9	2.27	5.7	.0707	6.5
Cr^{51}	6.60	52.8	14.4	30.9	N.R.	----	N.R.	----

SFP = Spent Fuel Pool
RCS = Reactor Coolant System
WHT = Waste Holding Tank
NR = Not Reported

7.0 Comparisons, Interpretations, and Recommendations

This study provides a preliminary data base on the radionuclide composition, and actual and relative concentrations to be found in the predominant waste forms generated by the four LWRs, which are processed, packaged and shipped to commercial radioactive waste burial sites. In addition, examination of the characteristics of the waste processing systems used to process and package the waste, and the analytical results provides an insight into the factors that need be considered in establishing a future expanded program of sampling and analysis.

7.1 Variables Influencing Composition of Waste Samples

The composition and relative radionuclide concentration in the samples of waste generated at LWRs is influenced by the following factors;

(a) Type of reactor and waste processing systems.

(b) Extent of release of fission products from failed fuel elements in the reactor core into the primary coolant (primarily a function of reactor operating time).

(c) Extent of corrosion products in the primary coolant (primarily a function of reactor operating time).

(d) Type of waste form sampled (i.e., filter sludge, resins, or evaporator bottoms).

(e) Location in waste processing system sample is drawn from (e.g., in individual waste streams vs. mixture in collection tanks).

(f) Age of sample from time of initial generation of the waste to time of analysis (concentrations of radionuclides will change as a function of half lives).

In addition to the above noted factors, the ability to accurately determine the composition of the sample is a function of sample size, solids content, and analytic procedures followed in the laboratory.

7.2 Comparisons of Radionuclide Analyses

A study of LWR wastes would be most useful if the pattern of radionuclide concentrations could be ascertained for the types of waste examined, so that information could be developed about concentrations of the radionuclide in the processed waste shipped to the burial site.

An attempt to draw definitive conclusions from the data obtained under this program and from prior laboratory analyses was impossible due to the lack of a sufficient number of similar samples necessary to provide statistical accuracy, and due to a lack of information on the operating experience pertinent to the samples collected. However, analyses can be performed to determine preliminary trends from the selective examination of classes of radionuclides in specific waste forms.

The data from the evaporator concentrate and spent ion resin waste form was used for this comparison. The filter sludge analyses were not considered due to the variability in sample form, lack of information on sample history and wide range in reported analyses.

In the case of the evaporator concentrates the variables effecting the data are further limited by considering the gamma emitting radionuclides reported in the literature (See Section 6.0) for the two PWRs, R.E. Ginna and Indian Point No. 2, for which data was compiled under this program. In addition, the effect of variation in sample composition as a result of the differential decay of the radionuclide inventory in the period between generation of the waste and sample analysis is minimized by further limiting the comparison to those radionuclides having half lives greater than 300 days. With the restrictions, it is felt that direct comparison of the selected radionuclide concentrations can be made. Table 7-1 presents the radionuclide concentrations and relative proportion of total activity of the selected nuclides for Cs^{137}, Cs^{134}, Co^{60} and Mn^{54}.

A similar restricted comparison was then made of the evaporator concentrate analyses determined under this program for the two PWRs and two BWRs. This data is presented in Table 7-2.

In the case of the spent ion exchange resin, the same type of analysis was applied to all of the long lived gamma emitters reported in the sample analyses from both this program and all those reported in the literature. This data is presented in Table 7-3.

TABLE 7-1

Comparison of Concentrations of Gamma Emitting Radionuclides ($T_{\frac{1}{2}}$>300 days) In Samples of Evaporator Concentrate From PWRs

Radionuclide		Reactor			
		Indian Point No. 2		R.E. Ginna	
		Sample of 9/75	Sample of 3/76	Sample of 1975	Sample of 2/76
Cs^{137}	Concentration	0.169μCi/ml	0.300μCi/ml	0.542μCi/ml	0.102μCi/gm
	Relative Prop.	55.2%	54.0%	61.6%	64.2%
Cs^{134}		0.113	0.190	0.272	0.037
		36.9	34.2	30.1	23.2
Co^{60}		0.017	0.035	0.045	0.019
		5.6	6.3	5.1	11.9
Mn^{54}		0.007	0.031	0.021	0.001
		2.3	5.6	2.3	0.6
Total Concentration in Selected Sample		0.306	0.556	0.880	0.159

TABLE 7-2

Comparison of Concentrations of Gamma Emitting Radionuclides ($T_{\frac{1}{2}}$>300 days) in Samples of Evaporator Concentrate From PWRs & BWRs

Radionuclide		PWR		BWR	
		Indian Point No.2	R.E. Ginna	Nine Mile Point	J.A. Fitzpatrick
Cs^{137}	Concentration	0.300μCi/ml	0.102μCi/gm	0.229μCi/gm	0.0004μCi/ml
	Relative Prop.	54.0%	64.2%	44.3%	1.9%
Cs^{134}		0.190	0.037	0.169	0.0001
		34.2	23.2	32.7	0.5
Co^{60}		0.035	0.019	0.096	0.0089
		6.3	11.9	18.6	42.0
Mn^{54}		0.031	0.001	0.023	0.0118
		5.6	0.6	4.4	55.7
Total Concentration in Selected Sample		0.556	0.159	0.517	0.0212

TABLE 7-3

Comparison of Concentrations of Gamma Emitting Radionuclides
($T_{\frac{1}{2}}$>300 days) in Samples of Spent Ion Exchange Resins From PWRs & BWRs

Radionuclide		R.E. Ginna *	R.E. Ginna **	Indian Point No.2 *	Unidentified PWR *	Nine Mile Point **
Cs^{137}	Concentration	28.23μCi/ml	21.9μCi/ml	5.77Ci	1.43μCi/ml	31.7μCi/gm
	Relative Prop.	30.2%	6.0%	30.2%	54.2%	77.5%
Cs^{134}		7.38	12.4	1.31	0.39	2.9
		7.9	34	7.0	14.8	7.1
Co^{60}		37.29	2.06	11.0	0.52	6.24
		39.9	5.6	57.7	19.7	15.2
Mn^{54}		20.47	.16	1.0	0.30	0.09
		21.9	.4	5.2	11.4	.2

* Sample from other programs
** Sample from this program

7.3 Interpretations of Data

Interpretations can be made of the radionuclide analyses determined under this and prior programs, and comparisons made between selected portions of the data, with the proviso that these interpretations are of preliminary trends (or patterns) and certainly cannot be considered to be definitive. The following interpretations appear to be justifiable.

I. Evaporator Concentrates

(a) Of the three waste forms examined, the consistency of the sample sources and of identifiable patterns in the data permits more supportable conclusions to be drawn with regard to evaporator concentrate compositions.

(b) The comparison of the relative concentrations of long half lived gamma emitting radionuclides (see section 7.2) shows that, with the exception of the sample from Fitzpatrick, the relative proportion of the constituents appears to be essentially of the same order for each reactor sampled under this program; and for the PWRs (where data was available) essentially of the same order as a function of time. This may imply a pattern in the relative concentrations of all the radionuclides in the evaporator concentrate samples. This initial pattern should serve as a reference point for future more detailed studies.

(c) The predominant gamma emitting radionuclides present in evaporator concentrates from all the reactors, with the exception of the samples from Fitzpatrick, are Cs^{137}, Cs^{134}, Co^{60}, and Co^{58}, generally in that order. This agrees with the information provided in the literature (Ref. 4). On the basis of the half lives of the gamma emitters, Cs^{137}, Cs^{134}, Co^{60}, and Mn^{54} will generally be predominant in the buried waste. Furthermore, Fe^{55}, Ni^{63}, and H^{3}, because of their long half lives

must also be considered as potential major constituents of the buried waste. It is reiterated that significant concentrations of individual radionuclides in the analyzed samples are not necessarily indicative of the relative long term importance of the radionuclides in terms of release and migration potential.

(d) The lack of agreement between the radionuclide analysis in the sample from Fitzpatrick and the other reactors along with its significantly lower total activity may be attributed to the short period of system operation at Fitzpatrick. It would be anticipated that the contribution from corrosion and fission products would be minimal during the early stages of reactor operation. Thus, the majority of the radionuclides present are activation products, while at the older plants, fission products tend to predominant. This can be related to the greater integrity of the fuel cladding in the early phases of plant operation.

(e) The data from radionuclide analyses of evaporator concentrate samples taken over a period of months from Indian Point No. 2 (see section 6.2) show appreciable variations in actual and relative concentrations of the radionuclides which cannot be correlated with reactor operations.

II. Spent Ion Exchange Resins

(a) The results of the various radionuclide analyses reported herein are too inconsistent to permit any trends to be discussed in the actual or relative concentrations of radionuclides. The comparison of the relative concentrations of the long lived gamma emitting radionuclides (see section 7.2) does not show, as it did in the case of the evaporator concentrates, any repeatable pattern among the samples.

(b) The predominant radionuclides present in spent ion exchange resin samples from all of the reactors are Cs^{134}, Co^{60}, and Mn^{54}, which occur in varying proportions in each sample. Since these radionuclides are all relatively long lived ($T_{\frac{1}{2}}$>300 days), they will generally be predominant in the buried waste.

III Filter Sludges

(a) The results of the various radionuclide analyses reported herein are too inconsistent to permit any trends to be discerned in the actual or relative concentrations of radionuclides.

(b) The predominant radionuclides present in the samples of filter sludges or "equivalent" vary, but are inclusive of Cs^{137}, Cs^{134}, Co^{60}, Co^{58}, Co^{57}, Fe^{55}, and Mn^{54}.

7.4 Recommendations

The following is recommended with regards treatment of the results of this study and for future work.

(1) The radionuclide analyses and their interpretations reported herein should be considered as preliminary indicators of trends and should be used as a tool in establshing the parameters for a more definitive program.

(2) In any future program, the sampling program must permit collection of a sufficient number of samples having the same parameters so as to be statistically reliable. To achieve this, samples similar in waste form, duration of reactor operation, age since generation, and location within the waste system should be obtained. Samples of sufficient size must be taken to permit standard laboratory analyses to be made and reported in consistent units. The radionuclide analyses should cover the full spectrum of radionuclides present.

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APPENDIX A

A. Waste Treatment Systems at Reactors From Which Samples Were Collected

The following sections describe the liquid and solid radwaste systems in use at the four commercial nuclear power plants at the time the samples were collected. The participating facilities were the Nine Mile Point, James A. Fitzpatrick, R.E. Ginna, and Indian Point No. 2 nuclear power stations.

A.1 Nine Mile Point (BWR) [5]

A.1.1 Liquid Radwaste System

The liquid radwaste system at Nine Mile Point is subdivided into (1) the waste collector subsystem, (2) the floor drain subsystem and (3) the regenerant chemical subsystem. A diagram of the system is presented in Figure A-1.

The waste collector subsystem processes those potentially radioactive liquid wastes which are characteristic of low conductivity. The wastes collected by this subsystem includes liquid waste from the reactor cooling system, the condensate system , the feedwater system, the reactor water clean-up system, the condensate demineralizer regeneration system and waste evaporator distillate. Any radioactive materials in these wastes are removed by filtration and ion exchange. The processed liquids are either reprocessed or sent to the condensate storage tank for in-plant reuse. The filter sludge is processed by the solid radwaste system. The ion exchange filters are regenerated and the regeneration solutions are processed by the regenerant chemical subsystem.

The floor drain subsystem collects all potentially radioactive high conductivity waste liquids from floor drains, laboratory drains, radwaste building sumps and decontamination drains. The collected liquids are passed through filters and then through demineralizers. The filtrate is either recovered or discharged while the sludge is processed by the solid radwaste

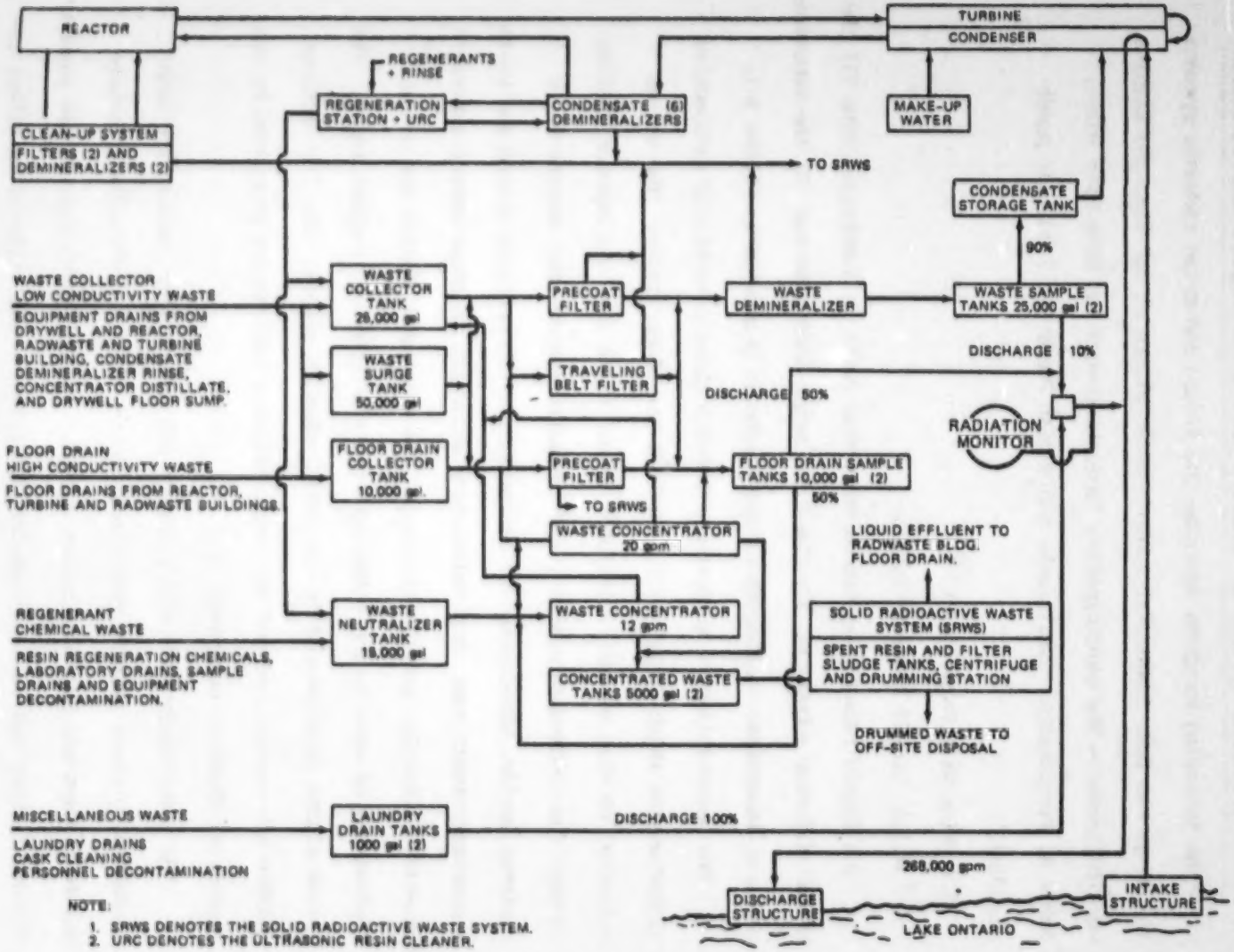
REACTOR
TURBINE
CONDENSER
REGENERANTS + RINSE
REGENERATION STATION + URC
CONDENSATE (6) DEMINERALIZERS
MAKE-UP WATER
CLEAN-UP SYSTEM
FILTERS (2) AND DEMINERALIZERS (2)
TO SRWS
CONDENSATE STORAGE TANK
90%
WASTE COLLECTOR LOW CONDUCTIVITY WASTE
EQUIPMENT DRAINS FROM DRYWELL AND REACTOR, RADWASTE AND TURBINE BUILDING, CONDENSATE DEMINERALIZER RINSE, CONCENTRATOR DISTILLATE, AND DRYWELL FLOOR SUMP.
WASTE COLLECTOR TANK 26,000 gal
WASTE SURGE TANK 50,000 gal
PRECOAT FILTER
TRAVELING BELT FILTER
WASTE DEMINERALIZER
WASTE SAMPLE TANKS 25,000 gal (2)
DISCHARGE 10%
DISCHARGE 50%
RADIATION MONITOR
FLOOR DRAIN HIGH CONDUCTIVITY WASTE
FLOOR DRAINS FROM REACTOR, TURBINE AND RADWASTE BUILDINGS.
FLOOR DRAIN COLLECTOR TANK 10,000 gal.
PRECOAT FILTER
TO SRWS
FLOOR DRAIN SAMPLE TANKS 10,000 gal (2)
50%
WASTE CONCENTRATOR 20 gpm
LIQUID EFFLUENT TO RADWASTE BLDG. FLOOR DRAIN.
REGENERANT CHEMICAL WASTE
RESIN REGENERATION CHEMICALS, LABORATORY DRAINS, SAMPLE DRAINS AND EQUIPMENT DECONTAMINATION.
WASTE NEUTRALIZER TANK 15,000 gal
WASTE CONCENTRATOR 12 gpm
SOLID RADIOACTIVE WASTE SYSTEM (SRWS)
SPENT RESIN AND FILTER SLUDGE TANKS, CENTRIFUGE AND DRUMMING STATION
CONCENTRATED WASTE TANKS 5000 gal (2)
DRUMMED WASTE TO OFF-SITE DISPOSAL
MISCELLANEOUS WASTE
LAUNDRY DRAINS
CASK CLEANING
PERSONNEL DECONTAMINATION
LAUNDRY DRAIN TANKS 1000 gal (2)
DISCHARGE 100%
268,000 gpm
DISCHARGE STRUCTURE
INTAKE STRUCTURE
LAKE ONTARIO
NOTE:
1. SRWS DENOTES THE SOLID RADIOACTIVE WASTE SYSTEM.
2. URC DENOTES THE ULTRASONIC RESIN CLEANER.

FIG. A-1 UPGRADED LIQUID RADWASTE SYSTEM, NINE MILE POINT NUCLEAR STATION, UNIT 1.

system,

The regenerant chemical subsystem collects those chemical wastes which result from the regeneration of the condensate demineralizers. These wastes are collected, neutralized and sampled in the waste neutralizer tank. From this tank the wastes are pumped to the waste evaporators, which are of 12 and 20 gpm capacity, where they are processed. The distillate is collected and is routed, eventually, to the waste collector subsystem. The waste concentrate is pumped to the solid radioactive waste system.

A.1.2 Solid Radwaste System

The wastes handled by this system include (1) evaporator concentrates, (2) filter sludges, (3) spent ion exchange resins, and (4) miscellaneous trash.

The evaporator concentrates are the solid wastes which remain from the processing of those wastes collected in the waste neutralizer tank and processed by the system's two waste concentrators.

The waste evaporator concentrates are routinely monitored in order to determine when the normal operational limit of 3μCi/ml is reached. Upon reaching the operational limit, the concentrate is pumped either to a concentrate waste tank from which it is subsequently pumped to the mixer or directly to the mixer where it is mixed with urea formaldehyde under the correct physio-chemical conditions. The mix is then pumped into a 150 cubic foot disposal Hittman liner for storage and subsequent transportation and burial.[6]

Filter sludges result from the filtration of those liquid wastes collected in the waste collector subsystem and floordrain subsystem. The filters are travelling belt-type filters which are designed to (1) reduce backwash water and (2) permit utilization of ultrasonic resin drains to remove resin crud

thus increasing the length of time between resin regeneration. In both systems the filter is designed to discharge a damp solid crud which is then handled by the solid waste system. This crud is incorporated with urea formaldehyde and the mix is pumped into the shipping cask for storage, transporation and burial.

Spent resins from the mixed bed demineralizer, are flushed directly to a 165 cubic foot capacity spent resin tank for storage. After a sufficient decay period has elapsed, or if more volume is required, the spent resins are pumped directly to the disposable Hittman shipping cask where they are dewatered prior to shipment. At the present time, solidification of the spent resin is being considered.[6]

A.2 James A. Fitzpatrick (BWR)

A.2.1 Liquid Radwaste System[7]

The wastes collected by the liquid radwaste system at Fitzpatrick are classified as high purity, low purity, chemical, detergent and sludge wastes. A flow chart of the liquid radwaste system showing the steps in processing each type of waste is provided in Figure A-2.

The high purity liquid wastes from the reactor coolant clean-up system, the residual heat removal system, waste and turbine buildings, are brought to the waste collector tank (30,000 gallons). The wastes are processed by filtration and demineralization. After processing, the filtrate is analyzed to determine whether the filtrate should be reused, reprocessed or discharged. The filters, filter sludges, and demineralizers are processed by the solid waste system.

Low purity liquid wastes, from the dry well, reactor, radwaste, and turbine building floor drains, are collected in a floor drain tank (8,500 gallon). These wastes are processed by filtration prior to transfer to one

of the floor drain sample tanks (17,000 gallons each). In these tanks the processed waste is sampled and subsequently analyzed. Based on the results of the analysis performed, these wastes are either discharged to the environment or subjected to additional processing in the chemical waste system or the high purity waste system.

The chemical wastes, collected from condensate demineralizer regeneration solutions, non-detergent decontamination and laboratory drains, are collected, neutralized, and sampled in one of the waste neutralizer tanks (17,000 gallons each). After sampling,these wastes are pH adjusted (7.0 to 9.0) prior to transfer to one of the two 20-gpm waste evaporators. The distillate from the evaporation process is sent to the waste collector tank (high purity waste system). The concentrate is either subject to further concentration in 0.8-gpm evaporator or sent directly to one of the two concentrate waste tanks.

The detergent waste system collects laundry, personnel decontamination and other detergent wash down wastes. These wastes are filtered prior to discharge. If activites higher than expected occur, the waste is transferred to the chemical waste system.

The waste sludge system is designed to collect waste filter, floor drain filter, and fuel-pool filter backwash and sludges in a filter sludge tank (11,000 gallons). The sludges are permitted to settle prior to decanting to the low purity waste system. Once decanted the sludge is transferred to the centrifuges for dewatering. The backwash from the reactor waste cleanup filter demineralizer precoat is collected in two phase separator tanks. The backwash is permitted to settle. The supernate is decanted to the high purity waste system and the sludges to the centrifuges before being sent to the solid waste system.

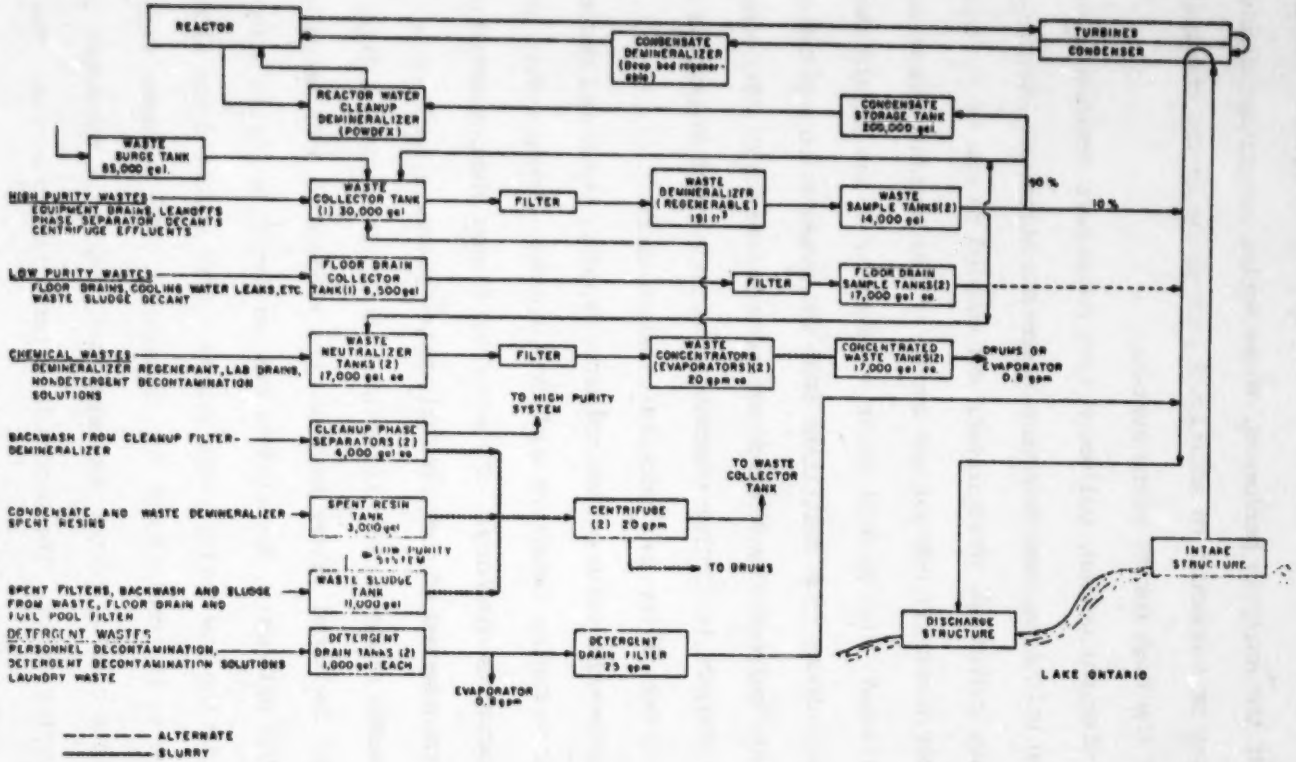
REACTOR
CONDENSATE DEMINERALIZER (Deep bed regenerable)
TURBINES
CONDENSER
REACTOR WATER CLEANUP DEMINERALIZER (POWDEX)
CONDENSATE STORAGE TANK 500,000 gal
WASTE SURGE TANK 85,000 gal.
HIGH PURITY WASTES
EQUIPMENT DRAINS, LEAKOFFS PHASE SEPARATOR DECANTS CENTRIFUGE EFFLUENTS
WASTE COLLECTOR TANK (1) 30,000 gal
FILTER
WASTE DEMINERALIZER (REGENERABLE) 150 ft³
WASTE SAMPLE TANKS(2) 14,000 gal
90 %
10 %
LOW PURITY WASTES
FLOOR DRAINS, COOLING WATER LEAKS, ETC. WASTE SLUDGE DECANT
FLOOR DRAIN COLLECTOR TANK(1) 8,500gal
FILTER
FLOOR DRAIN SAMPLE TANKS(2) 17,000 gal ea
CHEMICAL WASTES
DEMINERALIZER REGENERANT, LAB DRAINS, NONDETERGENT DECONTAMINATION SOLUTIONS
WASTE NEUTRALIZER TANKS (2) 17,000 gal ea
FILTER
WASTE CONCENTRATORS (EVAPORATORS)(2) 20 gpm ea
CONCENTRATED WASTE TANKS(2) 17,000 gal ea
DRUMS OR EVAPORATOR 0.8 gpm
TO HIGH PURITY SYSTEM
BACKWASH FROM CLEANUP FILTER-DEMINERALIZER
CLEANUP PHASE SEPARATORS (2) 4,000 gal ea
TO WASTE COLLECTOR TANK
CONDENSATE AND WASTE DEMINERALIZER SPENT RESINS
SPENT RESIN TANK 3,000gal
CENTRIFUGE (2) 20 gpm
TO DRUMS
LOW PURITY SYSTEM
SPENT FILTERS, BACKWASH AND SLUDGE FROM WASTE, FLOOR DRAIN AND FUEL POOL FILTER
WASTE SLUDGE TANK 11,000 gal
DETERGENT WASTES
PERSONNEL DECONTAMINATION, DETERGENT DECONTAMINATION SOLUTIONS LAUNDRY WASTE
DETERGENT DRAIN TANKS (2) 1,000 gal EACH
DETERGENT DRAIN FILTER 25 gpm
EVAPORATOR 0.8gpm
INTAKE STRUCTURE
DISCHARGE STRUCTURE
LAKE ONTARIO
ALTERNATE
SLURRY

Fig. A-2 Flow Chart of the Liquid Waste System.

James A. Fitzpatrick Nuclear Power Station

A.2.2 Solid Radwaste System [8]

The solid radwaste system at Fitzpatrick is divided into two subsections. The first subsection is designed to handle dry solid wastes (rags, paper, solid wastes, etc.) These wastes are compressed when possible in 55 gallon drums prior to transportation to a burial facicility.

The second subsection is designed to handle wet, solid wastes i.e., precoat materials, ion exchange resins and concentrate materials.

Precoat materials are discharged from filter-demineralizers into one of the two phase separator tanks. After settling has occurred the liquid is transferred to the waste collector tank for subsequent treatment and reuse. Precoat filters from waste, floordrains and fuel pool filters are discharged to the waste sludge tank. After permitting solids to settle the liquid is pumped to the floordrain sample tank. When the concentration of solids in the waste sludge tank reaches 1-5%, the concentrates are pumped to one of the two centrifuges and subsequently to the radwaste building for solidification.

Spent resins from the radioactive waste and condensate demineralizers are sluiced to a spent resin tank (3000 gallons) for storage prior to being fed to one of the two centrifuges (20 gpm). Spent resins are discharged directly from the centrifuges to the waste solidification facility.

A.3 R.E. Ginna (PWR)

A.3.1. Liquid Radwaste System [9]

All liquid wastes processed by this system whether collected by floor drains, equipment drains, laboratory drains or personnel decontamination drains are brought to the Waste Holdup Tank. A generalized schematic of the liquid system is shown in Figure A-3.

These collected liquids are then transferred to the evaporator feed tank from which they are pumped into the evaporator. The contents of the evaporator and the evaporator feed tank are circulated together and sampled every 4 hours. This analysis is conducted to determine when the operational limit of 2μCi/ml or 10% boric acid concentration is reached.[10] Once the concentrate reaches either of these limits, it is pumped to the solid waste system.

The distillate from the evaporation process is pumped first to the distillate tank and then to a waste condensate tank where it is analyzed and its release rate calculated.

A.3.2 Solid Radwaste System[9]

The solid radioactive waste generated at R.E. Ginna is composed primarily of evaporator concentrates and spent ion exchange resins.

The evaporator concentrates are pumped from the evaporator feed tank to the drumming station where the concentrate is mixed into vermiculite-cement mixture in 55-gallon drums. These drums are then moved to the drum storage area to await transportation to the disposal site.

The majority of the primary coolant system demineralizers are not designed to be regenerated. Under routine operating conditions, the spent resins are replaced by flushing and new resins installed.

The flushed resin is transferred to the spent resin storage tank where it remains until sufficient decay has occurred or more storage room is required. The flushed resin is then pumped to the drumming station where it is dewatered and placed in a 100 cubic foot Atcor shipping cask.

Any regenerant solutions, from the regeneration of the polishing demineralizers, are pumped to the waste holdup tank and then processed by the waste evaporator.

A.4 Indian Point 2 (PWR)

A.4.1 Liquid Radwaste System [11]

The liquid radioactive waste processing system at Indian Point-1 was being used to handle the liquid radwaste produced by Indian Point-2 at the time the samples were collected. The liquid radwaste handling system is designed to collect, treat, process and store all potentially radioactive liquid wastes generated on-site.

The collection center for these liquids consists of four waste collection tanks. The collected waste is subsequently transferred to the waste gas stripper. The removed waste gases are vented to the waste gas condenser and then processed by the gaseous waste system. The stripped liquid waste is pumped to the waste evaporator by means of an evaporator feed pump system.

The distillate from the evaporation process is passed through a polishing waste demineralizer and collected in the waste distillate storage tank. The collected distillate is sampled and, depending on the activity levels, is either transferred to the clear water storage tank or discharged to the environment. The concentrate is pumped to a sludge storage tank where it is held until transferred to the solidification processing facility.

The liquid waste handling facility at Indian Point Station is currently being improved. In the improved system the waste, after initial waste gas stripping, will be passed through filters into a feed pre-heater. From the pre-heater, the waste will be passed through a second gas stripper. After gas-stripping, the waste will be processed by two larger capacity evaporators. The concentrate will be pumped directly to the solidification station. The distillate will be passed through an absorption tower and

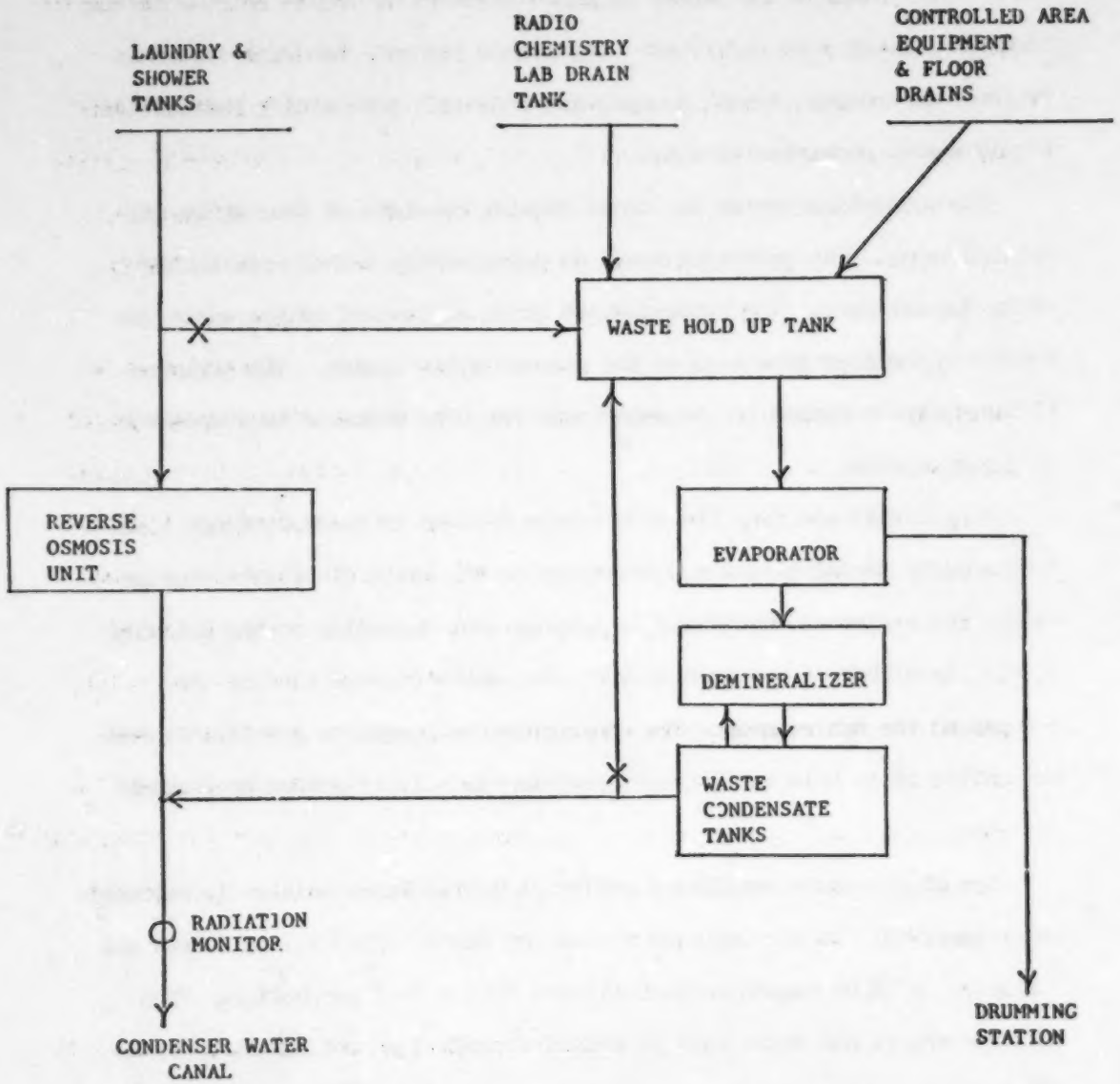

FIGURE A-3

LIQUID WASTE SYSTEM AT R.E. GINNA

a distillate cooler and then to two large volume distillate tanks. Operation of this new system has been initiated with the exception of the distillate storage tanks.[12]

The chemical and volume control system at Indian Point 2 is functional and is designed to handle and process reactor coolant letdown water.

The coolant letdown water passes through both regenerative and non-regenerative heat exchangers and a mixed bed coolant filter before storage in a volume control tank. From this control tank the coolant water is either pumped directly into the reactor coolant system or indirectly, by injection, into the seals of the reactor coolant pumps.

Liquid effluents from the reactor coolant system, containing boric acid, are collected in hold-up tanks for the purpose of recovering boric acid and reactor make-up water. Liquid from the hold-up tanks is passed through the evaporator feed ion exchanger, and the ion exchange filter before entering the waste gas stripper. The effluent from the stripper is transferred to the boric acid evaporator where the dilute boric acid is concentrated. The gases from the evaporator are condensed and cooled, passed through an evaporator condensate demineralizer and filter, and collected in a monitor tank. From this tank, the condensate is pumped to the primary water storage tank. The evaporator concentrates are discharged through a concentrate filter and into a concentrate holding tank before transfer to a boric acid tank.

A. 4.2 Solid Radwaste System

The predominant portion of the waste handled by this system results from the treatment and processing of liquid, radioactive wastes. These wastes are essentially evaporator concentrates, spent resins, filter sludges and filters.

Evaporator concentrates from the liquid radwaste systems are collected and mixed with urea formaldeyde and a catalyst in either 35 or 55 gallon drums. The future use of a 1500 gallon cask is under consideration.[12]

There are no regenerable resins in the liquid radwaste system or the chemical and volume control system, except for the boric acid evaporator condensate demineralizer. All resins are sluiced to a spent resin storage tank where they are held until sufficient decay has occurred. They are then pumped to the solidification facility where they are mixed with urea formaldehyde and a catalyst in a 200 ft^3 Atco cask.

Filters and filter sludges are handled in a manner similar to the way in which the concentrates are handled. These filters and sludges are (1) reactor coolant filters, (2) seal water filters, (3) seal injection filters, (4) spent fuel pool filters, (5) ion exchange filters, and (6) boric acid filters.

APPENDIX B

B. Analytical Methods Used By the Radiological Science Laboratory (RSL) [16]

B.1 Sample Preparation

Measurements of evaporator concentrate samples were performed on a dilution of each sample. Most of the other samples were fused with NaOH and the melt dissolved in distilled water. Two sludge samples were dissolved in acid, but portions of these samples were fused with NaOH for the C^{14} and I^{129} measurements.

B.2 Gross Alpha/Gross Beta Analysis

An aliquot of a water sample or fusion extract was evaporated and the residue quantitatively transferred to a planchet. The sample planchets were counted on a gas flow proportional counter. Sample planchets which required only gross-beta analysis were covered with saran wrap and counted on the alpha/beta plateau. Sample planchets which required both gross-beta and gross-alpha analyses were left uncovered and counted first on the alpha/beta plateau then on the alpha plateau.

The method is only adequate for screening purposes. Loss of volatile radionuclides, such as radioiodine and tritium, is one problem. Another drawback is the difficulty in radiometric standardization for a mixture of unknown alpha and beta emitters.

The radionuclides used as standards in the Radiological Science Laboratory are:

a. For gross beta - Sr^{90}-Y^{90}

b. For gross alpha - natural uranium

B.3 Gross Alpha - Spectrometric

A small aliquot of the liquid sample was evaporated to dryness and digested with nitric acid. After electrodeposition from an ammonium suplhate solution onto a stainless steel disc, the radioactivity was measured with a silicon surface-barrier detector, using only the counting efficiency to calculate the activity on the disc at each energy region. The plating efficiency for specific radionuclides was not known, so the method served only to qualitatively identify α-emitters present in the sample.

B.4 Isotopic-Uranium Analysis

U^{232} was added as a tracer to determine chemical and electrodeposition recovery. Water samples and fusion extracts were evaporated to dryness then taken up in 7.2 $\underline{N}$ HNO_3. Some sludge samples were leached with aqua regia extracting plutonium, uranium, americium, cerium and iron. Plutonium was collected from the leach solution by a batch ion exchange process, leaving uranium and iron in the leach solution, which was then evaporated to dryness. The residue from the pre-treatment and evaporation of the sample was dissolved in 7.2 $\underline{N}$ HNO_3 .

Uranium and any remaining plutonium were oxidized to the (IV) valence state with sodium nitrite. The plutonium nitrate complex formed in the strong nitric acid solution was removed on an anion exchange column. The effluent was evaporated to dryness, taken up in 9 $\underline{N}$ HCl and the uranium chloride complex adsorbed on an anion exchange column. Iron was removed from the column with a solution of 9 $\underline{N}$ HCl - 0.25 $\underline{M}$ NH_4I. The uranium was then eluted with 1.2 $\underline{N}$ HCl and electroplated onto a stainless steel disc from an ammonium sulphate solution.

The electroplated disc samples were counted on an alpha spectrometry system using a 450 mm^2 silicon surface barrier detector. The

system amplifier was biased to cover an energy range of about 4 Mev to 6 Mev.

The net cpm in each region were calculated and the values corrected for interference from higher energy alpha peaks, if necessary. The U^{235} and U^{238} activity levels were then calculated by applying the appropriate chemical recovery and counting efficiency factors.

B.5 Isotopic-Plutonium Analysis

Pu^{242} was added as a tracer to determine chemical and electrodeposition recovery. Water samples and fusion extracts were evaporated to dryness, then taken up in the 7.2 N HNO_3. Some sludge samples were leached with aqua regia, plutonium collected from the leach solution by a batch ion exchange process, then eluted and the eluate evaporated to dryness.

The residue from the pre-treatment of the sample was dissolved in 7.2 N HNO_3. Plutonium was oxidized to the (IV) valence state with sodium nitrite and the plutonium nitrate complex formed in the strong nitric acid solution was absorbed on an anion-exchange column. The column was washed with HNO_3 and HCl solutions, then the plutonium was eluted with a 0.36 N HCL - 0.01 N HF solution. Plutonium was electrodeposited from an ammonium sulphate solution onto a stainless steel disc.

The electroplated disc samples were counted on an alpha-spectrometry system using a 450 mm^2 silicon surface-barrier detector. The system amplifier was biased to cover an energy range of about 4 Mev to 6 Mev.

The net cpm in each region were calculated and the values corrected for interference from higher energy alpha peaks, if necessary. The Pu^{238} and $Pu^{239,240}$ activity were then calculated by applying the appropriate chemical recovery and counting efficiency factors.

B.6 Am^{241} Analysis

Am^{243} was added as a tracer to determine chemical and electrodeposition recovery. Water samples and fusion extracts were evaporated to dryness, then taken up in 7.2 N HNO_3. Some sludge samples were leached with aqua regia extracting plutonium, uranium, americium, curium, and iron. Plutonium was collected from solution by a batch ion exchange process leaving uranium, americium, curium and iron in the eluent which was then evaporated to dryness. The residue from the pre-treatment of the sample was dissolved in 7.2 N HNO_3.

Uranium and any remaining plutonium were oxidized to the (IV) valence state with sodium nitrite. The plutonium nitrate complex formed in the strong nitric acid solution was removed on an anion exchange column. The effluent was evaporated to dryness, taken up in 9 N HCl and the uranium chloride complex adsorbed on an anion exchange column. The effluent was collected for separation of americium. Iron was removed from the column with a solution of 9 N HCl - 0.25 M NH_4I, and the solution was combined with the effluent just previous to be used for the americium separation. The combined solution was evaporated to dryness, oxidize iodine with concentrate HNO_3, and the residue dissolved in 0.5 N HCl.

Americium was separated from the solution on a cation exchange resin, Dowex 50 x 8(H^+). The column was washed with 0.5N HCl and the americium eluted with 12 N HCl. The eluent was taken to dryness and americium was electroplated from ammonium sulphate solution onto a stainless steel disc.

The electroplated disc samples were counted on an alpha-spectrometry system using a 450 mm^2 silicon surface-barrier detector. The system amplifier was biased to cover an energy range of about 4 Mev to 6 Mev.

The net cpm in each region was calculated. The Am^{241} activity was then calculated applying the appropriate chemical recovery and counting efficiency factors.

B.7 Analysis of Tritium as HTO

Samples were vacuum distilled and the distillate collected to separate tritium from other interfering radionuclides and to remove chemical and/or physical quenching agents. An aliquot of the distillate was mixed with an organic scintillator and counted in a liquid scintillation spectrometer. Water known to be of low tritium content was used as a background sample.

The degree of quenching in a sample was determined by external standardization. The quench factor obtained was used to determine the counting efficiency for calculation of the tritium activity in the sample. Analysis of a 10 ml aliquot of the distillate resulted in a sensitivity of approximately 500 pCi/l.

B.8 Isotopic Gamma Analysis Ge(Li)

The liquid or solid sample in a standardized geometry, was analyzed with a Ge(Li) detector system. The system utilized a 4096-channel analyzer with an energy calibration of 0.5 keV/channel.

The activity of each gamma-emitting radionuclide in the sample was determined by using the efficiency factor for the photopeak of the isotope. The efficiency was obtained from a gamma-ray efficiency curve, prepared by measuring selected standards , in the standardized geometry, and using their known gamma ray intensities to determine photon efficiencies.

B.9 Sr^{90} Analysis

Sr^{85} tracer and stable strontium were added to the sample. The Sr^{85} tracer was used to radiometrically determine the chemical recovery of

strontium, while the stable strontium acted as a carrier. Water samples and fusion extracts were acid digested and strontium precipitated as the carbonate. Some sludge samples were dried and strontium removed by leaching twice with 6 N HNO_3. The leach solutions were evaporated to dryness and the residue taken up in HCl. Iron was removed on an anion exchange column and strontium in the effluent was precipitated as the oxalate then converted to the oxide.

The carbonate or the oxide from the sample pretreatment was dissolved in nitric acid. The rare earths, ruthenium and any remaining calcium was removed by precipitation of strontium nitrate from concentrated HNO_3. Yttrium carrier was added and the sample set aside 10-14 days for Y^{90} ingrowth.

At the end of the ingrowth period, yttrium was precipitated as the hydroxide, purified by repeated extractions into TBP and back-extractions into water. Yttrium was collected as the hydroxide, reprecipitated as the oxalate, converted to the oxide and mounted in a filter paper disc. The yttrium recovery was determined gravimetrically. The yttrium oxide was mounted on a nylon planchet and counted in an end-window, gas-flow proportional counter.

The chemical recovery for strontium was determined by gamma counting the Sr^{85} tracer on a NaI detector.

Three or more measurements, beginning immediately after the chemical separation of yttrium from strontium and continuing at approximately 2-day intervals, were made on the Y^{90} fraction in order to follow its decay. A computer program, using the half-life of Y^{90} as a known, performed a least-squares-fit to the counting data to calculate the Sr^{90} activity.

B. 10 Radioiodine Analysis

Stable iodine carrier was added to the sample to determine chemical recovery. Samples were treated to convert all iodine in the sample to a common oxidation state prior to chemical separation and purification.

Water samples were taken through an oxidation-reduction step using hydroxylamine hydrochloride and sodium bisulfite to convert all iodine to iodide suitable for processing through an anion exchange column.

Sludges were fused with a $NaOH-Na_2CO_3$ mixture. The melt was cooled, dissolved in distilled water and sodium hypochlorite added to oxidize the iodine to iodate. Hydroxylamine hydrochloride then reduced the iodate to elemental iodine for CCl_4 extraction.

After samples had been treated to convert all iodine in the sample to a common oxidation state, the iodine was isolated by solvent extraction or a combination of ion exchange and solvent extraction steps.

Iodine, as the iodide, was concentrated by adsorption on an anion exchange column. Following a NaCl wash, the iodine was eluted with sodium hypochlorite. Iodine, as iodate, was reduced to elemental iodine for extraction as palladium iodine.

Chemical recovery of the added carrier was determined gravemetrically.

The PdI_2 precipitate was counted on an intrinsic-germanium detector and the intensities of the Kα X-rays from Te^{125} and Xe^{129} measured.

The decay of I^{131} also results in the production of xenon X-rays. Consequently I^{131} constituted an interference in the procedure. Prior to the X-ray measurement, all samples were counted for 100 minutes on a NaI well-detector to check for the presence of I^{131}. A second measurement on the intrinsic diode after two weeks decay provided further verification of I^{131}. If I^{131} was present, the X-ray data was corrected for I^{131} interference or the sample allowed to decay until the I^{131} activity no

longer seriously interfered with the I^{129} measurement.

The germanium detector is standardized for both I^{125} and I^{129} as a function of weight of the PdI_2 precipitate. The Kα X-rays at 27.5 and 27.2 keV for Te^{125} and 29.7 and 29.4 keV for Xe^{129} are used to quantitate the data. The matrix coefficients to correct for the interference of one spectral region to the other are also determined from the standard spectra for I^{125} and I^{129}. Correction factors for I^{131} interference are determined from I_2Pd^{131} standards.

The counts in the I^{125} region and the I^{129} region were summed separately. The net counting rate in each region was computed. A matrix calculation was used to correct the I^{125} net counting rate and the I^{129} net counting rate for mutual interference from Compton interactions and I^{131} interference. The appropriate decay, volume, counting efficiency and chemical recovery corrections were then applied to compute the I^{125} and I^{129} activities.

B.11 Te^{99} Analysis

Technetium was separated by solvent extraction with nitrobenzene. Stable rhenium was added to the sample to determine the chemical recovery . The rhenium was oxidized to the perrhenate and technetium was oxidized to the pertechnetate. An extraction was performed from dilute nitric acid into nitrobenzene, using tetraphenyarsonium chloride as the extracting agent. The pertechnetate and perrhenate were then back-extracted into concentrated nitric acid. Tetraphenylarsonium-pertechnetate and perrhenate were then reprecipitated. The precipitate was filtered and the rhenium recovery is determined gravimetrically. Te^{99} was counted in an end-window gas-flow-proportional counter.

B.12 C^{14} Analysis

Sludge and resin samples were first fused with NaOH and the resulting melt dissolved in distilled water. Water samples were analyzed directly.

The extraction of CO_2 and CH_4 was carried out in a closed vacuum system. A sample volume of 50-100 ml was spiked with 0.1 g of sodium carbonate, introduced into the vacuum system, and 50 ml of concentrated hydrochloric acid was added under vacuum. The sample was constantly purged with He containing a total of 25 ml (STP) of methane carrier gas. The evolved CO_2 and the stripped methane were then collected and separated cryogenically after removal of the water vapor in a series of cold traps. Subsequently, the gases were purified in a gas chromatograph and the extraction yield determined volumetrically. The purified gas was loaded into an internal gas-proportional counter and diluted in the counter with P-10 counting gas. Spectral analysis was performed by pulse-height analysis under controlled conditions in a massive iron shield, where the counting tube was operated inside an anticoincidence guard counter.

B. 13 Fe^{55} Analysis

Stable iron was added as a carrier to determine chemical recovery. Water samples, fusion extracts, and acid leachates were evaporated to dryness and the residue dissolved in a 50% acetone-water solution. The sample was then passed through a chromatographic column containing AG50W-X8 cation-exchange resin which had been equilibrated with 50% acetone-water solution. The iron (III) was eluted with 80% acetone-0.5 M HCl solution. Iron was electrodeposited from a $NH_4H_2PO_4$-$(NH_4)_2CO_3$ solution onto a polished copper disc, and the 5.9 keV X-ray was then measured with an instrinsic-germanium detector.

B. 14 Ni^{63} Analysis

Nickel was isolated from water samples, fusion extracts and acid leachates by forming nickel dimethylglyoximate which was extracted into

chloroform. Nickel carrier, measured spectrophotometrically, was used to determine the chemical recovery. The nickel dimethylgloximate was decolorized with hydrochloric acid and the 67-keV beta of Ni^{63} counted on a liquid scintillation spectrometer.

B.15 Detection Limits

The detection limits varied for each sample measurement inasmuch as these limits are a function of the quantity of sample used, counting time, and processing recovery, which varied. The detection limits of radionuclides measured by isotopic gamma analyses also vary with the gamma composition of the sample. The deviations on the measured samples ranged from $\pm$ 5% to greater than $\pm$ 80%, without any consistent pattern for individual radionuclides.

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